

## NUCLEAR DATA FOR U-Pu FUEL CYCLE EVALUATION

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Abstract :

The need for nuclear data for the U-Pu fuel cycle evaluation is reviewed for both thermal and fast reactors. It is interesting to note that the higher burn-up rates planned for future LMFBR's or already obtained for PWR have not so far significantly modified the requests lists.

The various methods used to derive the requests are also described. Some of them, which recently appeared, allow the use of integral data for the assessment of microscopic data requests. This is one way to stress the importance of the coupling of the microscopic and integral information.

The progress made during the past 10 years on the evaluation tools and associated parameter data bases is impressive. It results from the accumulation of microscopic data of high quality and from the incorporation of more and more basic physics into the models.

The test of the evaluated files against integral data requires several conditions :

- quality in the data processing,
- cleanness of the experimental environment,
- correctness of the neutronic calculations.

Such conditions are considered to have been met in consistent sets of integral data which have been used to check major data files in the thermal and the fast ranges concerning the Minacs, FP, structural materials. The quality of the evaluated data available for the U-Pu fuel cycle valuation is satisfying in most cases. The quality of the evaluation tools presently available is a guarantee that the last major requests will be satisfied within a reasonable time scale provided that a minimum of well targeted microscopic measurements are performed.

The long standing problem of the production of  $^{236}\text{Pu}$  via the  $(n,2n)$  process on  $^{237}\text{Np}$  is described in detail as an excellent example, possibly to be followed in the future, by that which can be obtained when looking at the possible convergence of the information from various sources : integral, microscopic, nuclear model.

The present status of the nuclear data for U-Pu fuel cycle has been analysed following the various progresses made during the past 10 years.

I. THE NEED AND THE REQUESTS - SHORT REVIEW

The requests have to be divided into 2 different categories.

Category I

Those requests related to clearly specified and quantified needs which are urgent since they are connected with feasibility or economic aspects. They concern all the in pile fuel cycle demands and a part of the out of pile cycle demands. The requested accuracies on the nuclear parameters concerned are quantified, in principle.

Category II

Those requests which are related to needs only defined in a qualitative way because they correspond to the long term or because they are still not of practical concern.

In this case one wishes only to derive an error bar on the integral parameter from the presently known accuracies on the nuclear data.

The need comes from the problems encountered during the irradiated fuel transportation, manipulation or reprocessing, namely :

Safety (criticality control), biological shielding, decay heat, are the constraints of the out-of-core cycle. Some of the related needs belong to the category I.

Those which may affect the reactor performances, namely :

The loss of reactivity per cycle decay heat, inherent neutron source, are the constraints of the in core fuel cycle. All the related needs belong to the category I.

The fuel cycle problems will be reviewed according to the following distribution :

1. Loss of reactivity per cycle

It is important to have a correct knowledge of the fuel composition as a function of time to adequately manage the fuel during the reactor operation and predict the effective length of the cycle, to define strategies for fuel recycling or actinide incineration.

The typical target accuracy on reactivity loss/cycle is  $0.1 \div 0.3 \% \Delta K/K$  in  $\frac{\Delta K}{K}$ , distributed between Minor Actinides (Minac's) and Fission Product (FP) according to their relative contribution.

The requested accuracies on Minac's for both LWR's and FBR's are summarized in a review by J. BOUCHARD / 4 / and have to be slightly modified to account for studies since 1979.

The request on the global contribution of the FP is  $\pm 5 \%$  to be distributed in the following way, assuming a partial correlation of the items / 5 / :

$$\pm 3 \% \text{ for } \sigma_{n,\gamma}$$

$$\pm 20 \% \text{ for } \sigma_{n,n'} \text{ (estimated)}$$

+ 2,5 % for the fission yields

+ 2 % for the migration effect of the volatile products

## 2. Neutron emission

The major sources are :

The spontaneous fission and the  $(\alpha, n)$  reactions in the case of oxide fuels.

. Inherent source (for FBR's) :

Neutron control and reactivity measurements in subcritical configurations are of concern.

The target accuracy on the inherent source is : + 10 % and the major uncertainties are related to :

$^{242}\text{Cm}$ ,  $^{244}\text{Cm}$  ( $\nu_{\text{sf}}$ ,  $\lambda_{\text{sf}}$ ) and  $(\alpha, n)$  cross sections for the isotopes  $^{17}\text{O}$ ,  $^{18}\text{O}$  and impurities like N or F. The Pu-240 spontaneous fission data are sufficiently well known.

. Biological shieldings :

For transport flasks of irradiated fuel, the target accuracy is + 20 % for both LWR's and FBR's.  $^{242}\text{Cm}$ ,  $^{244}\text{Cm}$  are concerned, together with the nuclei from which they originate ;

i.e. :  $^{241}\text{Pu}(\lambda, \sigma_{\text{n}, \text{f}})$ ,  $^{241}\text{Am}(\text{n}, \gamma)$ ,  $^{242}\text{Cm}(\sigma_{\text{n}, \text{f}})$ .

## 3. $\alpha$ , $\beta$ , $\gamma$ radioactivity of the fuel

The related problems are on :

- the decay heat (reactor shutdown accidental or not, irradiated subassemblies transport, long term storage of the wastes),  
- the biological shielding in the reprocessing plants or transport flasks.

The need is 5 % - 10 % for both LWR's and FBR's on the total residual heat and its components :  $(\beta + \gamma)$  decay of FP's,  $(\beta + \gamma)$  decay related with  $^{239}\text{U}$  decay,  $\alpha$  decay of the actinides.

The associated uncertainties are on :

- F.P. yields,  
- average  $\beta, \gamma$  energies and spectra,  
- decay constants.

Concerning the biological shielding, the requests are on the atom density :

+ 5 % for  $^{241}\text{Am}$ , + 20 % for  $^{238}\text{Pu}$ , + 30 % for  $^{236}\text{Pu}$ .

## 4. Radioactive nuclei production

This results from the activation of the coolant itself and impurities, of the coolant circuits and components.

The needs are of category II, and concern especially the following nuclei and reactions :

$^{50}\text{Cr}(\text{n}, \gamma)^{51}\text{Cr}$ ,  $^{54}\text{Fe}(\text{n}, \alpha)^{51}\text{Cr}$ ,  $^{54}\text{Fe}(\text{n}, \text{p})^{54}\text{Mn}$ ,  $^{58}\text{Fe}(\text{n}, \gamma)^{59}\text{Fe}$ ,  $^{58}\text{Ni}(\text{n}, \text{p})^{58}\text{Co}$ ,  $^{60}\text{Ni}(\text{n}, \text{p})^{60}\text{Co}$ ,  $^{59}\text{Co}(\text{n}, \gamma)^{60}\text{Co}$  and also  $^{40}\text{Ar}(\text{n}, \gamma)^{41}\text{Ar}$ ,  $^{36}\text{Ar}(\text{n}, \gamma)^{37}\text{Ar}$ ,  $^{23}\text{Na}(\text{n}, \text{p})^{23}\text{Ne}$ ,  $^{23}\text{Na}(\text{n}, \gamma)^{24}\text{Na}$ ,  $^{23}\text{Na}(\text{n}, 2\text{n})^{22}\text{Na}$ , these last two reactions being also contributors to the decay heat of FBR'S.

## 5. Gas production

The need seems to be limited to the  $^3\text{H}$  production and is related to the environment safety.

There are two sources :

- reaction  $^{10}\text{B}(\text{n}, 2\alpha)\text{T}$ ,  
- ternary fission of the heavy nuclei.  
The request is + 10 % on the production.

## 6. New needs

Apart a special request on the fission yields of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  (requested accuracy 1.5 %) for irradiated PWR fuel subassembly identification, the main new request concerns the inelastic scattering cross section of the FP's, especially the even-even isotopes with an estimated requested accuracy of + 20 %. This results from the significant contribution of this cross section to the reactivity effect of the FP's, recently stressed by PALMIOTTI and SALVATORES / 6 / and H. GRUPPELAAR et al / 7 /.

It has to be noted that the new conditions resulting from Pu recycling in PWR's (MOX fuel) and increased burn-up rates (PWR's, FBR's of the future generation) policy have not been so far reflected in the request list. Normally one can expect better accuracy requirements for  $^{236}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{243}\text{Am}$  and to a major extent for  $^{244}\text{Cm}$ .

For the moment, the criticality problems (LWR's) are more dependent on calculational schemes or methods and are solved in a conservative way. In the future, needs for neutron data (Gd, Hf, Eu, thermal cross sections and

resonance integrals,  $^{242}\text{Cm}(\nu_{\text{sf}}, \lambda_{\text{sf}})^{244}\text{Cm}(\nu_{\text{sf}}, \lambda_{\text{sf}})$  will probably appear.

According to a recent Japanese study, the burn-up characteristics of HCLWR are strongly affected by self-shielding effects of the fission products. Needs for an accurate determination of data in the resolved range are likely, if HCLWR are built on an industrial scale.

Problems with actinide incineration may be highlighted again. In that case one can imagine requests for neutron data ( $\sigma_{\text{f}}, \sigma_{\text{g}}, \nu$ ) together with fission yields in both thermal and fast ranges for higher minor actinides.

The requests are derived through a multi-step procedure :

(a) Assessment of the need.

(b) Identification of the nuclei and nuclear constants from inventory codes and sensitivity calculations.

(c) Determination of the requested accuracy using techniques based on the perturbation theory and variational methods.

$$\frac{\Delta P^2}{P^2} = \sum_k \sum_i S_{ki}^P \frac{\text{var}(p_{ki})}{P_{ki}^2} + \sum_K \sum_i \sum_{j \neq i} S_{ki}^P S_{kj}^P \frac{\text{cov}(p_{ki}, p_{kj})}{P_{ki} \cdot P_{kj}} + \sum_k \sum_i \sum_{j \neq k} \sum_\ell S_{ki}^P S_{j\ell}^P \frac{\text{cov}(p_{ki}, p_{j\ell})}{P_{ki} \cdot P_{j\ell}} \quad (1)$$

In this expression  $k, j$  stand for the isotope,  $i, l$  for the particular item of the isotope  $k$  or  $j$ , while  $\Delta P$  refer to the requested absolute uncertainty on the parameter  $P$ .

A typical example of this method is given by PATRICK and SOWERBY / 1 /.

Usachev has suggested treating the needs together by defining a "cost" function :

$$F = \frac{W_j}{\Delta P_{kj}^2} \text{ to be minimized in the frame of}$$

conditions, each of them being associated to a specific problem :

$$\Delta P^2 > \sum_k \sum_i (S_{ki}^P \Delta P_{ki})^2 \quad (2)$$

It is important to stress the fact that new methods appeared recently which derive the requested uncertainties on nuclear constants by taking into account information already available from integral experiments. In that case (2) becomes :

$$\sum_j \sum_l S_j^k S_l^k v_{jl} < DP^2 \quad (3)$$

That is the case for a method proposed by BOBKOV, PYATNITSKAYA and USACHEV / 2 / who define a "penalty" function  $\Phi$  consisting of the function  $F$  plus a factor  $1/r$  times the conditional constraints,  $r$  being a parameter.  $\Phi$  has to be solved unconditionally.

The method presented by GANDINI and SALVATORES / 3 / for the statistical consistent adjustment can be extended to the accuracy requirements. A likelihood function  $L$  is constructed on a vector containing the requested and already obtained accuracies of both microscopic and integral parameters and on a dispersion matrix  $B$  based on the covariance matrices for the microscopic and integral data. The solution is obtained when one finds one estimator of  $\Delta P$  which maximises  $L$ .

$$L = C^{te} \exp - \frac{1}{2} \left\{ \left( \frac{\Delta P}{P} \right)^T B^{-1} \left( \frac{\Delta P}{P} \right) \right\} \quad (4)$$

These methods which are tools to assess requirements on microscopic data by accounting for integral information suggest the following comments :

- the integral and microscopic information compete to define an optimum program of measurement,
- they are complementary to define a status of knowledge.

They suggest also that the evaluation procedures should systematically include integral information.

A general consensus exists for the requests of category I and II for the needs considered together. When differences appear they can be understood as resulting from different priorities attributed in the national programs in the fields of economy or safety and that involves the point (a).

## II. REVIEW OF THE EVALUATION TOOLS AND DATA BASE

The improvement of the evaluation tools and associated parameter data bases are very impressive. They result from few new measu-

rements of high quality on one side, from the incorporation of more basic physics into the phenomenological models on the other.

Two papers by J.R. SALVY / 9 / and M. GARDNER / 10 / have reviewed the advances made in the past 10 years in the calculational methods. Both papers are complementary.

The main points of these improvements are as follows :

The general problem of the Optical model parameterization has been seriously investigated by MADLAND and YOUNG / 8 / and J. SALVY / 9 / among many others. The Optical model parameterization has to obey to the general constraint of reproducing the low energy parameters (Neutron Strength functions for "S" and "P" waves, scattering radius. The so called "SPRT" method / 11 / has been defined for that purpose) and the direct interaction effect resulting from a strong deformation of the nucleus. P.G. YOUNG / 12 / recommends that the regional or local parameterization be preferred to the global spherical parameterization derived for large mass ranges unless they are locally appropriate. These general prescriptions were observed in the actinide evaluation. On the contrary, the systematic use of the global spherical optical model parameterization resulted in FP libraries of insufficient quality, especially concerning the inelastic cross section. Such situation has been pointed out by H. GRUPELAAR et al / 7 /. They explained the systematic disagreement on  $\frac{E}{C}$  values between results obtained in irradiation experiments on one side and reactivity worth measurements on the other for the even-even FP of the 100 mass region by a strong underestimation of the inelastic cross section.

The improvement of the systematics of the main parameters of the statistical model have to be highlighted. These improvements are clearly consecutive to improved techniques to derive them from the primary data (7.5 % on  $\langle D \rangle$ , 2 % on  $\langle \Gamma_n \rangle$ ) / 13 /.

They concern :

- The level density parameter "a". Phenomenological systematics are still of interest, but a better definition of the pairing energy will, hopefully, bring additional improvements as noted by IIJIMA et al. / 14 /, ROHR et al / 15 / and many others. Nevertheless better physics will be helpful and BCS calculations, as an example, would give information on the energy dependence of "a".

- Collective effects should be taken into consideration in the relationships to describe the level densities.

- The importance of non statistical effects is stressed in the gamma decay of the compound nuclens. The  $\gamma$  strength functions ( $E1, M1, E2$ ) are considered as more reliable to predict the penetrabilities provided that appropriate normalization constraints are put in adequate energy regions. The systematics on these functions are preferred to those on  $\langle \Gamma \gamma \rangle$  and  $\langle D \rangle$  made independently / 10 /.

- The importance of the spectroscopic description by modeled discrete levels in the first few MeV above the ground state has been repeatedly justified by GARDNER / 10 / not only for the cross section but also for the isomeric

rate calculations. The condition is on a sufficiently large number of generated levels so as the spin J and K number distributions be statistically significant.

- The systematics by J. BEHRENS / 16 / on fission cross sections in terms of constant excitation energy have reached a high degree of confidence (10 %) over a large energy range of the fast domain (0.6 MeV - 20 MeV). That is the result of an extensive set of coherent measurements.

For lower energies one has to combine the systematics on more basic parameters such as barrier heights, curvature parameters, fission channel densities to be considered altogether. The systematics given by J.E. LYNN / 17 / are useful but they have to be combined consistently with the more phenomenological systematics here above mentioned or those bringing complementary information on the average fission cross section in the plateau as a function of the fissibility parameter / 17 /.

The use of the pre-equilibrium formalism is unavoidable to describe the threshold reaction cross sections. In a review paper P.E. HODGSON and co-workers / 19 / have shown that most available codes are able to correctly describe the angular and energetic distributions of secondary particles, whatever the basis. Some progress is to be made for the (n,p) and (n, $\alpha$ ) reactions with small cross sections. For the (n,2n) or (n,3n) cross sections, especially just above the threshold important differences may appear if the  $\gamma$  competition or the level densities, especially in the discrete range, are not accurately described.

### III. VALIDATION ON INTEGRAL EXPERIMENTS

The test of the evaluated files against integral data requires several conditions : quality of the data processing, cleanness of the experimental environment, correctness of the neutronic calculations.

Such conditions are achieved in the resonance integral measurements, in clean experiments in critical facilities ( $K_{\infty}$ , reaction rate ratios etc), in the analyses of pure sample irradiation. Accuracies of 1.5 % - 3 % are common in the (n, $\gamma$ ) or (n,f) cross section tests and 10-50 % can be obtained for the (n,2n) cross section tests.

The post-irradiation analyses also satisfy the hereabove criteria but are less accurate and have to be handled very cautiously. 5 % at most can be obtained on the nucleus production or disparition cross sections. All these techniques have been widely used to validate, in the thermal and fast ranges, major files like KEDAK4, ENDFB4/5 in the recent past and JEF1 which is presently the object of a systematic and exhaustive effort.

Actinides :

Thermal and epithermal ranges.

H. KUSTERS / 20 / reports that only  $I_{\gamma}$  for  $^{243}\text{Am}$  still remains as a unsolved problem

$$\left(\frac{E-C}{C} \sim 20 \%\right).$$

Acceptable performances of JEF1 isotopes ( $^{240,241,242}\text{Pu}$ ,  $^{241,243}\text{Am}$ ,  $^{244}\text{Cm}$ ) have been suspected in the analysis of the irradiation SHERWOOD experiment / 21 /. Subassembly calculations for the "ICARE S" experiment bring

interesting preliminary information in the epithermal range for the capture cross sections of  $^{238}\text{U}$ ,  $^{239,240,241}\text{Pu}$  (See table 1).

TABLE 1 (taken from / 44 /)  
C/E VALUES FOR THE ICARE S EXPERIMENT USING JEF1 DATA

Type of data	C/E
$\sigma_c$ ( $\text{U-238}$ ) ( $\text{UO}_2$ )	$1.056 \pm 4.8 \%$
$\sigma_c$ ( $\text{Pu-239}$ ) (0.1 % Pu)	$1.02 \pm 4.4 \%$
$\sigma_c$ ( $\text{Pu-240}$ ) (0.1 %)	$0.971 \pm 4.4 \%$
$\sigma_c$ ( $\text{Pu-240}$ ) (0.5 %)	$1.038 \pm 4.6 \%$
$\sigma_c$ ( $\text{Pu-241}$ ) (0.1 %)	$0.907 \pm 4.2 \%$
$\sigma_c$ ( $\text{Pu-242}$ ) (0.1 %)	$0.903 \pm 7.6 \%$

Large size experimental programs of post-irradiation analyses have been initiated especially in FRANCE and Federal Republic of GERMANY. They resulted in synthetic information concerning PWR standard fuel at different burn-up rates (the analyses of high enrichment and MOX type fuel have not yet been finalized). Information has been obtained for the contents of  $^{235,236}\text{U}$ ,  $^{236,238,239,240,241,242}\text{Pu}$ ,  $^{241,243}\text{Am}$ ,  $^{242,244}\text{Cm}$  in OBRIGHEIM or TRINO2 VERCELLESE irradiated fuels by FISCHER and WIESE / 22 / using KEDAK4 and by MATEEVA and KUSTERS / 23 / using JEF1. KEDAK4 appeared in good shape while JEF1 showed some deficiencies for  $^{240,241}\text{Pu}$ .

Similar results have been obtained by GAUCHER and SANTAMARINA / 24 / by analyses of TIHANGE and FESSENHEIM fuels. Higher burn-up rates on one side and sensitivity studies on the other allowed more precise conclusions on

$\sigma_c$  of  $^{238}\text{U}$  (decrease of  $\Gamma_{\gamma}$  of the 3 first resonances), on  $\sigma_{n,2n}$  of  $^{238}\text{U}$ , on the isomeric

ratio  $\frac{^{242}\text{Am}^m}{^{242}\text{Am}^g}$ . Concerning the  $^{243}\text{Am}$  resonance

integral the conclusions are identical to KUSTERS.

These results are consistent with "ICARE S" ones.

Fast range :

Reaction rate measurements performed in ZEBRA / 25 / and MASURCA / 26 / have been used to check the Actinide data from UKNDL, KEDAK4 and JEF1 origin. C/E values close to 1 have been found and it is worth mentioning that most of the evaluations were obtained by theoretical calculations.

The informations obtained by pure sample irradiation in critical facility KNK II / 27 / or in power plant (PHENIX) / 28 / are probably the most valuable. They indicate that KEDAK IV and JEF1 capture data are in general good shape with indication for a reduction of  $\sigma_c$  for the higher Pu isotopes (see table 2).

TABLE 2 (taken from / 28 /)

TYPE OF DATA		C/E values with JEF1
		Average value (PROFIL-1 + PROFIL-2)
$\sigma_c$	(U-235)	0.97 ± 1.4 %
$\sigma_c$	(U-238)	0.96 ± 1.6 %
$\sigma_c$	(Pu-238)	0.95 ± 3 %
$\sigma_c$	(Pu-239)	0.97 ± 1.8 %
$\sigma_{n,2n}$	(Pu-239)	1.38 ± 11 %
$\sigma_{n,2n}$	(Pu-240)	1.06 ± 1.6 %
$\sigma_{n,2n}$	(Pu-240)	0.83 ± 14 %
$\sigma_{n,2n}$	(Pu-241)	1.11 ± 3.7 %
$\sigma_c$	(Pu-242)	1.16 ± 3.5 %
$\sigma_c$	(Am-241)	1.03 ± 1.4 %
$\sigma_c$	(Am-243)	0.94 ± 5 %

Using sensitivity calculation indications, D'ANGELO et al. have modified the branching ratios for  $^{241}\text{Am}$  and  $^{242}\text{Am}^g$  so as to obtain consistent results for the production of  $^{242}\text{Am}$ ,  $^{242}\text{Cm}$ ,  $^{242}\text{Pu}$  and  $^{238}\text{Pu}$ , indicating that the branching ratio of  $^{241}\text{Am}$  is still a problem to be solved.

By an additional analysis of PROFIL II experiment using JEF1 as data base, F. CLERI and A. D'ANGELO / 29 / confirmed the solution of the long standing problem of the production of  $^{236}\text{Pu}$  via the (n,2n) process on  $^{237}\text{Np}$ . They obtained a  $\frac{C}{E}$  value equal to  $1.19 \pm 0.15$  for the (n,2n) cross section to be associated to a value of 0.23 for the probability of decay on the short lived state  $6^-$  of  $^{236}\text{Np}$ . This last value is in accordance with theoretical calculations by GARDNER, GARDNER and HOFF / 30 /.

Post-irradiation analyses have brought significant informations. That is the case for PFR fuel / 31 / for 3 burn-up rate values, also for KNK II fuel / 31 /. But once again a special mention should be made of the TRAPU experiment for the high accuracy of its results. It consists in an irradiation in PHENIX of special fuel with different enrichments in high Pu isotopes. The results on  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  and higher Pu are consistent with the information from the irradiation experiments (See table 3).

TABLE 3 (taken from / 28 /)  
(E-C)/C VALUES ON FINAL (END OF IRRADIATION) COMPOSITIONS  
IN THE TRAPU EXPERIMENTS (PERCENTAGE VALUES) USING JEF-1 DATA

U-238 = 100	TRAPU 1	TRAPU 2	TRAPU 3
U-234	2.2 ± 2.5	0.2 ± 1.3	- 3.4 ± 1.0
U-235	0.8 ± 0.3	- 1.2 ± 0.2	- 0.9 ± 0.2
U-236	2.0 ± 0.5	0.0 ± 0.4	0.3 ± 0.3
Np-237	9.7 ± 6.8	11.3 ± 3.3	17.9 ± 3.2
Pu-238	- 1.8 ± 0.9	0.1 ± 0.4	0.9 ± 0.4
Pu-239	0.0 ± 0.4	1.6 ± 0.3	1.7 ± 0.3
Pu-240	0.6 ± 0.4	2.5 ± 0.3	1.6 ± 0.3
Pu-241	- 3.1 ± 0.4	0.3 ± 0.3	- 1.8 ± 0.3
Pu-242	- 7.4 ± 0.5	- 2.9 ± 0.4	- 1.3 ± 0.3
Am-241	5.2 ± 3.0	3.7 ± 3.6	3.4 ± 2.1
Am-242	- 26.5 ± 3.6	- 29.2 ± 4.0	- 26.6 ± 2.5
Am-243	- 7.8 ± 3.6	- 5.2 ± 4.0	- 7.7 ± 2.5
Cm-242	4.0 ± 2.4	5.4 ± 2.6	6.0 ± 2.1
Cm-243	-	- 11.4 ± 2.7	- 11.4 ± 2.6
Cm-244	- 2.9 ± 2.0	- 12.9 ± 2.2	- 13.8 ± 1.7

Fission Products :

Because of their major contribution to the reactivity loss per cycle in a power reactor (75 % for a FBR), a worldwide effort has been devoted to validate the microscopic data, essentially those involved in fast reactors. This effort has been applied to the separate isotopes through complementary methods such as reactivity worth measurements for different spectral indices (STEK measurements) or activation-transmutation rate measurements (CFRMF or PHENIX).

As already described at SANTA-FE Conference the situation of the capture cross section of the individual isotopes is quite acceptable except for a few isotopes. The strong underestimation of the inelastic cross section in most files has been clearly identified as responsible of the systematic discrepancy between reactivity worth and irradiation results. The even-even isotopes potentially candidates for a static rotational or dynamic vibrational deformation are concerned.

TABLE 4 (taken from / 7 /)  
E/C-VALUES IN FAST REACTOR SPECTRA

P <sup>+</sup>	Nuclide	STEK-1000+	CFRMF	RONA-3	PHENIX
1	Pd-105	1.02 ± .01			1.17 ± .04
2	Ru-101	.95 ± .02			.89 ± .03
3	Rh-103	1.01 ± .01			
4	Tc-99	1.05 ± .01	.80 ± .12		
5	Cs-133	1.05 ± .01	.99 ± .07		1.01 ± .03
6	Pd-107	1.02 ± .01			
7	Sm-149	1.04 ± .01			1.00 ± .02
8	Pm-147	1.00 ± .02	.81 ± .10		
9	Sm-151	.97 ± .01			1.20 ± .06
10	Mo-97	.88 ± .01			1.06 ± .03
11	Nd-145	1.03 ± .03			.89 ± .03
12	Nd-143	.99 ± .04			
13	Ru-102	(.67 ± .08)	.89 ± .06	.94 ± .04	
14	Ag-109	.98 ± .03	1.10 ± .11		
15	Eu-153	.98 ± .03	1.02 ± .07		
16	Mo-95	.89 ± .01			.96 ± .02
17	Ru-104	(.64 ± .11)	.96 ± .06	1.05 ± .05	
18	Xe-131	1.11 ± .07			
19	Mo-98	(.54 ± .03)	.82 ± .05	.73 ± .04	
20	Pd-108	(.99 ± .12)	1.10 ± .07	1.12 ± .07	
21	Pr-141	.91 ± .01	.97 ± .15	.96 ± .05	
22	Mo-100	(.78 ± .06)	1.12 ± .19	1.08 ± .06	
23	I-129	.97 ± .08	.94 ± .06		
24	Cs-135				
25	Eu-155				
26	Pd-106	(1.04 ± .11)			1.65 ± .11
27	Ru-103				
28	Xe-132		.91 ± .07		
29	Sm-152	1.08 ± .03	.92 ± .06	1.17 ± .06	
30	Gd-157				
31	Sm-147	1.00 ± .02			
32	Zr-93	.68 ± .11			
33	Xe-134		.57 ± .04		
34	I-127	1.05 ± .03	.90 ± .09		
35	Nd-146	(.72 ± .12)	.97 ± .06	.97 ± .05	
36	Nd-148	(.83 ± .07)	.85 ± .12	1.02 ± .05	
37	Ru-106				
38	La-139		.96 ± .05		
39	Nb-95				
40	Zr-96				

According to GRUPPELAAR and al. / 7 / the discrepancy is almost entirely removed by replacing the evaluated data by recent experimental data, like those of KONOBEVSKII and POPOV / 33 / or those of A.B. SMITH, GUENTHER and WHALEN / 34 /.

Concerning the global reactivity effect of FP, measurements made on irradiated fuel indicate the relevance of the following effects :

- The presence of nodules observed in the analysed fuel which, in addition to a correct selfshielding calculation, raises the problem of their origin for modelisation purposes.

- The migration of volatile FP whose effect to the global effect is about 6 %. (A special measurement of the axial distribution of  $^{133}\text{Cs}$  and  $^{135}\text{Cs}$  on an irradiated fuel pin has been made recently in FRANCE / 45 / to check the modelisation).

However, despite the fact that a complete analysis has not been made, the reported results indicate that the target accuracy ( $\pm 5\%$ ) on the global FP reactivity effect can be achieved, if the inelastic cross section problem is solved.

The problem of the neutron source are largely advanced since the situation of spontaneous fission has been clarified by an international coordinated effort (AIEA CRP). V. BENZI et al / 35 / have offered an interesting system including a data set, a theoretical model for  $^{17}\text{O}(\alpha, n)$  and  $^{18}\text{O}(\alpha, n)$  cross sections, and a calculational method. Such a system which was successfully checked against semi-integral data has been used to analyse subcritical neutron counting rates in SUPER-PHENIX1, and  $\frac{C}{E}$  values of 0.96 have been obtained / 36 /.

The integral validation of the  $\beta, \gamma$  decay data of the radioactive individual FP is made through fission pulse measurements. A review on that topics by J.K. DICKENS / 37 / indicates that the measurements do not exceed the time period concerned by the loss of coolant accident, ie  $10^4$ - $10^5$  sec, and are systematically underestimated by calculations. At short cooling time, when microscopic data are lacking, call is made to theoretical models to estimate the  $\beta, \gamma$  transitions and their strength. KRATZ / 39 / has produced a state of Arte" review of the theoretical models and systematics. Although not too much accurate the "Gross beta theory" has been largely used.

According to C.W. REICH / 38 / conclusions, the data of the nuclides close to the  $\beta$  stability valley (small  $Q_\beta$ , long half-lives) are satisfactory. Progresses are to be made for the nuclides more and more far from the  $\beta$  stability valley concerning half-life isomer ratio predictions.  $Q_\beta$  determination should be made by mass determination. All the remarks concern the short cooling times. For actual application good estimation of the fission pulse function can be obtained by a correct

determination of the systematic corrective factor to be applied to the summation calculations. An application of that procedure has been made by A. TOBIAS / 40 / to the JEF1 data library, with corrective factors everywhere less than 1.085 for  $^{235}\text{U}$  and  $^{239}\text{Pu}$ . A totally integral checking which involves more than the fission pulse ( $\alpha$  contribution,  $^{239}\text{U}$  decay, FP capture, neutronic calculations) is underway by analysing the decay heat measurement of the SUPER PHENIX power plant.

An important program of data validation for the major contributors to the steel activities has been developed in UK at ZEBRA mock-up / 41 /, in three different core regions of different enrichments and in breeder regions. Some reactions associated with the sodium coolant were also studied. The calculations were performed with the UK file and ENDFB5. The results give indication that for the most important reactions the data are in better shape than generally believed,  $\frac{C}{E}$  values between 0.8 and 1.2 being observed. These figures are perfectly acceptable for requests of category II. The details can be seen on the table 5.

The lack of data concerning the production of  $^3\text{H}$  by ternary fission on  $^{239}\text{Pu}$  has been filled by a very recent measurement at BORDEAUX University performed by BARREAU and OUSTI / 42 /, for the energy range thermal - 2 MeV.

The general trend observed for the  $^3\text{H}$  yield is slowly decreasing with the energy. A similar trend is observed for  $^{235}\text{U}$ . This result is in contradiction with the data by SHARMA / 43 /.

A typical example of a problem solved by looking at the convergence of informations from various sources

It concerns the long standing problem of the production of  $^{236}\text{Pu}$  via the (n,2n) reaction on  $^{237}\text{Np}$ . The production of  $^{236}\text{Pu}$  results from the following reaction scheme.

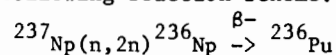


TABLE 5 (taken from / 41 /)  
COMPARISON OF C/E VALUES FOR MORE IMPORTANT STEEL ACTIVATIONS

Neutron Reaction	Fissile zones					Fertile zones		
	Core 14		BZB/3	BZD/3		Core 14		BZD/3
	Element	Steel	Steel	Element	Steel	Element	Element	Steel
Cr50(n $\gamma$ )Cr51	10.80 $\pm$ 6 %	10.87 $\pm$ 11 %	0.78 $\pm$ 6 %	0.80 $\pm$ 5 %	10.82 $\pm$ 6 %	10.91 $\pm$ 9 %	10.79 $\pm$ 6 %	10.79 $\pm$ 7 %
Fe54(n $\rho$ )Mn54	10.77 $\pm$ 5 % (0.91)	10.87 $\pm$ 11 % (1.03)	0.91 $\pm$ 6 % (1.07)	0.83 $\pm$ 4 % (0.97)	10.88 $\pm$ 5 % (1.04)	10.79 $\pm$ 11 % (0.94)	10.83 $\pm$ 6 % (1.00)	10.87 $\pm$ 7 % (1.05)
Ni58(n $\rho$ )Co58	10.96 $\pm$ 6 % (0.88)	11.10 $\pm$ 11 % (1.01)	1.07 $\pm$ 6 % (0.98)	1.02 $\pm$ 4 % (0.93)	11.10 $\pm$ 5 % (1.00)	-	11.04 $\pm$ 6 % (0.96)	11.10 $\pm$ 7 % (1.01)
Fe58(n $\gamma$ )Fe59	10.82 $\pm$ 6 % (0.99)	10.80 $\pm$ 11 % (0.96)	0.86 $\pm$ 6 % (1.02)	0.87 $\pm$ 5 % (1.04)	10.84 $\pm$ 5 % (1.00)	11.28 $\pm$ 11 % (1.26)	11.18 $\pm$ 6 % (1.24)	11.16 $\pm$ 7 % (1.22)
Co59(n $\gamma$ )Co60	10.80 $\pm$ 5 %	-	-	0.87 $\pm$ 5 %	-	11.16 $\pm$ 9 %	11.15 $\pm$ 6 %	-
Ni60(n $\rho$ )Co60	10.88 $\pm$ 11 % (0.97)	-	-	0.84 $\pm$ 5 % (0.93)	-	-	10.63 $\pm$ 12 % (0.69)	-

The  $\frac{C}{E}$  ratios in brackets use the ENDF B-V cross-sections

The short lived state  $6^-$  of  $^{236}\text{Np}$  at 0.077 MeV of excitation energy decays with an half-life of 22.5 h into  $^{236}\text{Pu}$  (48 %) and  $^{236}\text{U}$  (52 %).

The cross-section of  $^{236}\text{Pu}$  production can be written as :

$$\sigma_{^{236}\text{Pu}} = \sigma_{n,2n} \times 0.48 \times \frac{m}{m+g} \quad (4)$$

where  $\sigma_{n,2n}$  stands for the  $n,2n$  cross-section of  $^{237}\text{Np}$ , and  $\frac{m}{g}$  is the isomeric ratio of  $^{236}\text{Np}$ .

There are microscopic experimental data of  $^{236}\text{Pu}$  at low energy (threshold = 6.8 MeV - 9.6 MeV) by KORNILOV et al recently obtained and several older sets of data around 14 MeV which are consistent. Although the values just above the threshold have the most important weight in the production in reactor, a complete curve up to 14 MeV is necessary for a correct calculation of the production of  $^{236}\text{Pu}$ .

An evaluation is obligatory to prove that all the data sets are consistent and to interpolate between 9.6 MeV and 14 MeV. It should concern  $\sigma_{n,2n}$  and the isomeric ratio in the same footing or these two items separately. The evaluation of GARDNER and GARDNER of the first type proved to be too high by 30 % - 40 %. The evaluation performed at CADARACHE was limited to  $\sigma_{n,2n}$  only.

There are no experimental data for  $\sigma_{n,2n}$  and its evaluation is relevant to theoretical calculations.

The extrapolation of the first chance fission above the second chance fission threshold is a key point in such calculations. Proportion values of the second chance fission as a function of energy were derived by FREHAUT from vp measurement and have been used to check the calculation. The first convergence of microscopic data and model calculations has been found there.

The isomeric ratio as calculated by GARDNER et al by generating numerous discrete levels in the frame of basic physics constraints, show, using (4) :

- a good consistency with the experimental value at 14 MeV obtained by MEYERS,
- the calculated  $\sigma_{n,2n}$  is consistent with

the experimental value of  $\sigma_{^{236}\text{Pu}}$ . That is another convergence of microscopic data and model calculations,

- the data below 9.6 MeV are consistent with those at 14 MeV and the interpolation scheme is fixed together by the calculated  $\sigma_{n,2n}$  and the isomeric ratio.

At this stage one can say that the calculated and the microscopic measured values show a perfect internal consistency.

There are three sets of integral data of  $^{236}\text{Pu}$  production obtained by different ways and in different environment configurations. The data reported by PAULSON and HENNELY were obtained in various environments ranging from  $\text{D}_2\text{O}$  reflectors to PWR. The data by WIESE et al

were derived from post irradiation analysis of PWR fuel only. In both cases  $^{237}\text{Np}$  is a by product of U fuel irradiation. Its production results from several channels whose relative importance have to be known accurately. Another difficulty related to the thermal range is the determination of the neutron flux and its evolution along the irradiation. The pure  $^{237}\text{Np}$  sample irradiated in the fast power reactor PHENIX is not submitted to the hereabove mentioned constraints and the experiment is nicer in the principle. But, all the integral experiment analyses are strongly dependent on a correct determination of the  $^{236}\text{Pu}$  formation via the channel  $^{237}\text{Np}(\gamma,n)$  for which the estimation of the  $\gamma$  flux is the most uncertain point.

It has been proved that the averaged  $\sigma_{^{236}\text{Pu}}$  derived by PAULSON and HENNELY is in error by a factor 2.

On the contrary the analyses by WIESE et al on one side and by CLERI and D'ANGELO on the other agree and show that the JEF1 values are slightly overestimated with respect to the integral data by about the same quantity ( $\frac{C}{E} \approx 1.2$ ). In addition, in the work by CLERI and D'ANGELO, the branching ratio to be asso-

ciated to the cross-section is  $\left(\frac{m}{m+g}\right)^{\text{adj}}$

$= 0.77 \times 0.48 = 0.369 \pm 0.024$  in good agreement with GARDNER'S curve averaged in a fission

spectrum  $\left(\frac{m}{m+g}\right)^{\text{c}} = 0.336$ . There are two

examples of an agreement between model calculations and integral data.

Finally this problem can be considered as solved in a satisfactory way in the frame of the requests, thanks to complementary informations from various sources : microscopic, integral and nuclear model.

#### CONCLUSION

The main conclusions of this review are :

- The MINAC isotopes are revealed to be in a general good shape. Most of the discrepancies observed 10 years ago have been removed. That is particularly true for the  $^{241,243}\text{Am}(n,\gamma)$  x-section (fast range),  $^{242}\text{Pu}(n,\gamma)$  x-section (fast range), the long standing problem of the production of  $^{236}\text{Pu}$  via the  $(n,2n)$  process on  $^{237}\text{Np}$ .

Some MINACs still need minor revision :  $^{243}\text{Am}(n,\gamma)$  resonance integral,  $^{234}\text{U}(n,\gamma)$  (thermal),  $^{241}\text{Pu}(n,\gamma)$  x-section (fast range),  $^{240-241}\text{Pu}$  fission x-section (fast range). More serious are the following discrepancies still remaining whose removal needs further theoretical or experimental investigation :

- $^{239}\text{Pu}(n,2n)$  x-section and the isomeric ratio of  $^{242}\text{Am}$  are typical examples.

Although the capture cross sections of the individual FP have been shown to be, in general, of good quality, all the major files were found to underestimate by about 10 % the FP component in the reactivity-loss per cycle in a large size LMFBR. The origin lies in a significant (factor 2) underestimation of the inelastic x-section as a result of using the spherical optical model. Further experimental investigation is certainly required. For the purpose of application the urgent effort could be limited to the most important FP, namely the even mass Ru, Pd, Sm isotopes.

- The problems of the neutron source are largely advanced since the situation of spontaneous fission has been clarified by an international coordinated effort and the suggested solution for the ( $\alpha, n$ ) component (oxyde fuel) is being tested with satisfactory preliminary results.

- The data involved in the decay heat are in an acceptable situation, at least those concerned by current applications (LOCA). For better predictions at short cooling times significant improvements of the theory are needed.

The quality of the major contributors to the steel activation has been demonstrated to be satisfactory. That may not be the case for the minor activation or corrosion products, as a result of a difficult feedback from the sparse integral measurements.

- Measurements for a better understanding of the fission process have resulted in a better knowledge of the Tritium production by tripartition ( $^{235}\text{U}$  and  $^{239}\text{Pu}$ ) and its evolution with the incident energy.

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