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DECAY HEAT IN FAST POWER REACTORS :
OBJECTIVES QUALIFYING THE COMPUTER CODE

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RESUME :

The decay heat calculation code PROTEE has been checked
against integral measurements and specific component measu-
rement. Of particular relevance are the PHENIX measurements,
which are presented here together with the calculation expe-
riment value comparison. A reasonnable agreement is found and a
slight tendency ($\sim$ 10 %) to over estimation of the PROTEE code
is indicated.

CONTENTS

1 - INTRODUCTION

Design studies for nuclear plants and other fuel cycle facilities
must solve a number of problems related to thermohydraulic phenomena
or to dose rate estimates and biological shielding.

In this context it is essential to characterize not only the entire
reactor but also each subassembly individually, according to the overall
decay power and the various radiation emissions after irradiation in a
fast breeder reactor.

The major radiation sources are unstable nuclides formed under
irradiation by fission or neutron capture in the fuel or in metal
structures.

Two types of radiation may be distinguished according to their
interaction with various materials : radiation with low penetrating
power (α, β) and highly penetrating radiation (γ, n, X).

Both types of radiation are concerned by the overall decay heat
studies, while biological shielding and dose rate calculation studies
involve only radiations of the second type.

2 - THE NATURE OF THE PROBLEM

The afterpower in spent fuel must be estimated both for project
studies and for operation of fast breeder reactors and their related
fuel cycle facilities.

The energy released after irradiation from radionuclides created
by fission or neutron capture in the fuel and the reactor structures
raises a number of problems that may be summarized in two areas :
decay heat removal and biological shielding design.

2.1 - Decay Heat

The problem of decay heat removal must be solved for all
fuel cycle facilities including the reactor in order to prevent damage
or destruction of the metal structures that constitute the barriers
between the fuel and its environment. Such accidents would result in
unacceptable contamination hazards or safety-related incidents.

A number of phases in the fuel cycle are particularly
sensitive to these problems :

> . In the reactor, immediately following an emergency shutdown (e.g.
> with failure of the normal core cooling system) which
> could result in damage to the fuel cladding tubes or to
> metal reactor structures.

> . For spent fuel subassembly transfer devices and transport
> casks, in order to prevent cladding failure or container
> damage.

> . In the reprocessing plant, where decay heat could cause
> overheating of the fuel solutions, resulting in evaporation
> and fouling of the facility.

> . At all stations where spent fuel subassemblies or radioac-
> tive waste materials are stored, where any damage to the
> stored objects or containers could present a serious conta-
> mination or criticality hazard.

2.2 - Biological shielding

This problem area involves penetrating radiation such as
γ rays and neutrons, for which suitable biological shielding must be
designed to protect personnel from the spent fuel. This problem arises
at all fuel cycle stations outside the reactor, for example in trans-
porting spent fuel elements, where significant technical and financial
considerations are involved in optimizing the shielding from the stand-
point of size and weight.

3 - OBJECTIVES

The purpose of this effort is to develop and qualify experimentally an advanced computer program to estimate the physical values necessary to solve the problems mentioned above. The program must be capable of evaluating the thermal power as well as the radiation spectrum and flux based on the following parameters :

- the mass and composition of the fresh fuel and the metal structures,

- reactor operating and refueling schedule,

- post-irradiation cooling time.

The major physical parameters to be calculated include the total thermal power, and the intensity and spectrum of penetrating radiation (γ, neutrons).

The program is qualified by overall or specific measurements on the reactor and on irradiation fuel samples.

4 - THE PROTEE CODE

4.1 - Major Decay Heat Components

After shutdown of a 1200 MWe SUPERPHENIX type fast breeder
reactor, the major decay power components and their relative importance
are shown after various cooling times in the following table.

TABLE 1

DECAY POWER COMPONENTS FOR A 1200 MWe FAST BREEDER REACTOR OPERATED FOR 2 YEARS PRIOR TO SHUTDOWN					
COMPONENTS	RELATIVE PERCENTAGE AFTER VARIOUS COOLING TIMES				
	0	1 day	1 month	1 year	6 years
1- Fission product $\beta + \gamma$ energy	93%	63%	73%	63%	36%
2- ^{239}U & ^{239}Np $\beta + \gamma$ energy	6.5	27	0	0	0
3- Heavy nuclide α energy	0.5	5.8	23	32	53
4- Structural steel $\beta + \gamma$ energy	0.2	1.6	4.5	5.5	11
5- ^{22}Na & ^{24}Na $\beta + \gamma$ energy	~ 0	2.6	~ 0	~ 0	~ 0
TOTAL POWER (MWth)	210	20.4	5	1.1	0.25

The dose rate measured 1 meter from a subassembly 24 hours
after irradiation in the same type of reactor is given below.

TABLE 2

ITEM	DOSE RATE AT 1m	
	γ radiation	neutrons
1200 MWe reactor subassembly	6×10^5 rem/hour	1 rem/hour

The shielding required to transport such an assembly after
5 months of cooling would be on the order of 20 cm of lead (γ shielding)
and 20 cm of borated compound (neutron shielding).

4.2 - Computation Methods and Basic Data

The PROTEE code developed by the CEA is designed to compute
the energy or the radiation spectrum for each of the five major
components listed in Table 1.

4.2.1 - Component No 1 : Fission Product $\beta + \gamma$ Energy Release

The code processes the "elementary fission" curves,
i.e. the γ spectrum or afterpower decay curves after a single fission
of the major nuclides (^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu)

The "elementary fission" curves currently used here
are "best estimates" from an analysis of available experimental findings
and on a number of theoretical estimates based on the energy released by
each fission product. The estimates were further refined by two theore-
tical considerations relative to the fission process :

- a 15 MeV standard value for the total $\beta + \gamma$ fission
  energy release (previously used values on the order
  of 9 MeV)[1],

- the decay power due to fast neutron fission of
  ^{239}Pu is lower than for ^{235}U ; this significant
  finding means that the much more numerous results
  for ^{235}U can be used to limit the range of values
  for ^{239}Pu.

These curves have been corrected for capture effects
on individual fission products. The estimated correction resulted in a
4% increase in the calculated fast breeder reactor decay power value
for cooling times ranging from 0 to 2 years.

4.2.2 – Component No 2 : ^{239}U and ^{239}Np $\beta + \gamma$ Energy Release

The essential calculation parameter is the production of ^{239}U and ^{239}Np from ^{238}U by neutron capture. The short half-life of these nuclides means that their contribution to the decay power is practically limited to short cooling periods (0 to 10 days). The basic data of importance here include the desintegration branching. The mean energy actually released by β radiation (less the neutrino component) and the γ energy.

4.2.3 – Component No3 : α Energy Release from Heavy Nuclei & Neutron Source

This component represents the energy released as α radiation from heavy nuclei and neutrons emitted from spontaneous fissions or (α,n) reactions involving ^{18}O in the uranium or plutonium oxide.

This requires an estimate of the concentration of the major actinides : in practice, 30 nuclides ranging from ^{232}U to ^{244}Cm are taken into account. After computing the reactor neutron spectrum and flux, the following must be determined :

- Daughter products obtained by neutron capture or radioactive disintegration (half-lives, branching, etc.),

- Microscopic cross sections (capture, fission, (n,2n), etc ..) from the CEA's "CARNAVAL IV" project calculation code set,

- α decay spectra,

- Neutron spectra and intensities for (α,n) reactions involving ^{18}O according to the α spectrum,

- Neutron spectra and intensities for spontaneous fissions.

4.2.4 - Component No 4 : Energy Released by Activation of Steel Structures

This energy component accounts for a significant fraction of the total decay power only after cooling for at least a year or more, but represents a certain problem for transport and storage of irradiated steel wastes.

Amoung the most important radioactive nuclides in the steel, from the standpoint of shielding problems, is ^{60}Co formed by neutron capture in ^{59}Co, of which the steel contains only about 0,3%. Long-term storage problems are also created by long half-life radio-nuclides such as ^{63}Ni (92 years), ^{59}Ni (10^5years) or ^{93}Mo (3000 years).

Two sorts of difficulties arise in this study. One involves estimating the neutron capture rate for ^{59}Co in a fast breeder reactor : this reaction is largely due to low-energy neutrons (approx. 130 eV) which are negligible in the reactor neutron balance. The second is to obtain adequate cross section data throughout the energy spectrum for the formation of ^{63}Ni, ^{59}Ni and ^{93}Mo.

4.2.5 - Component No 5 : Sodium Activation Energy

This is a minor component in the total decay power, accounting only for about 2.5% during the first few days after reactor shutdown. The cross sections and neutron reaction rates are well known for ^{24}Na and ^{22}Na formation, as are the disintegration families and energy release values.

5 - QUALIFICATION

The PROTEE code is qualified primarily by comparison with experimental results in two basic areas : integral measurements covering the core or individual subassemblies, and specific measurements for each of the decay heat components.

The uncertainty bounds on the PROTEE values are estimated from the overall results of these comparisons.

5.1 - Integral Measurements

5.1.1 - Phenix Reactor

The total decay power was measured in the Phenix fast breeder reactor. After a normal reactor shutdown, the test involves cooling times ranging from 7 to 35 hours. The measurement procedure can be summarized as follows.

With the reactor shut down, the thermal balance involves only a limited number of parameters :

- the decay power to be determined $P_R(t)$,

- thermal outleakage from the facility $F(t)$,

- possible inputs $A(t)$ (pumping power, electric preheating, etc ..),

- thermal inertia in the facility I,

- temperature variations throughout the facility $\frac{d\theta}{dt}(t)$

The thermal balance at the instant (t) is expressed :

$$P_R(t) = I \frac{d\theta}{dt} + F(t) - A(t) \qquad (1)$$

Steps were taken to minimize both outleakage $F(t)$ and inputs $A(t)$ in order to ensure accurate measurements : the steam generator panels were closed, and the primary and secondary pumps were run at 300 rpm (this speed represents a trade-off between low thermal input and adequate temperature uniformity).

The temperature variation was limited to between 250°C and 290°C for reasons of accuracy and plant operating requirements. The measurement periods were separated by forced cooling intervals.

Sodium temperature variations $\theta(t)$ were monitored at 12 points in the reactor :

- at the inlets of the three primary sodium pumps

- at the inlets of 3 of the 6 heat exchangers

} Primary sodium

- at the inlets of 3 heat exchangers

- at the outlets of 3 heat exchangers

} Secondary sodium

The measured results showed that the sodium was practically isothermal.

The I and $F(t)$ values were measured during reactor startup testing, $A(t)$ is well evaluated ; given the $\theta(t)$ measurement data it was therefore possible to determine $P_R(t)$ from equation (1).

Figure 1 compares the computed and experimental results, and warrants a number of remarks.

The computed values are consistently about 10% higher than the measured findings.
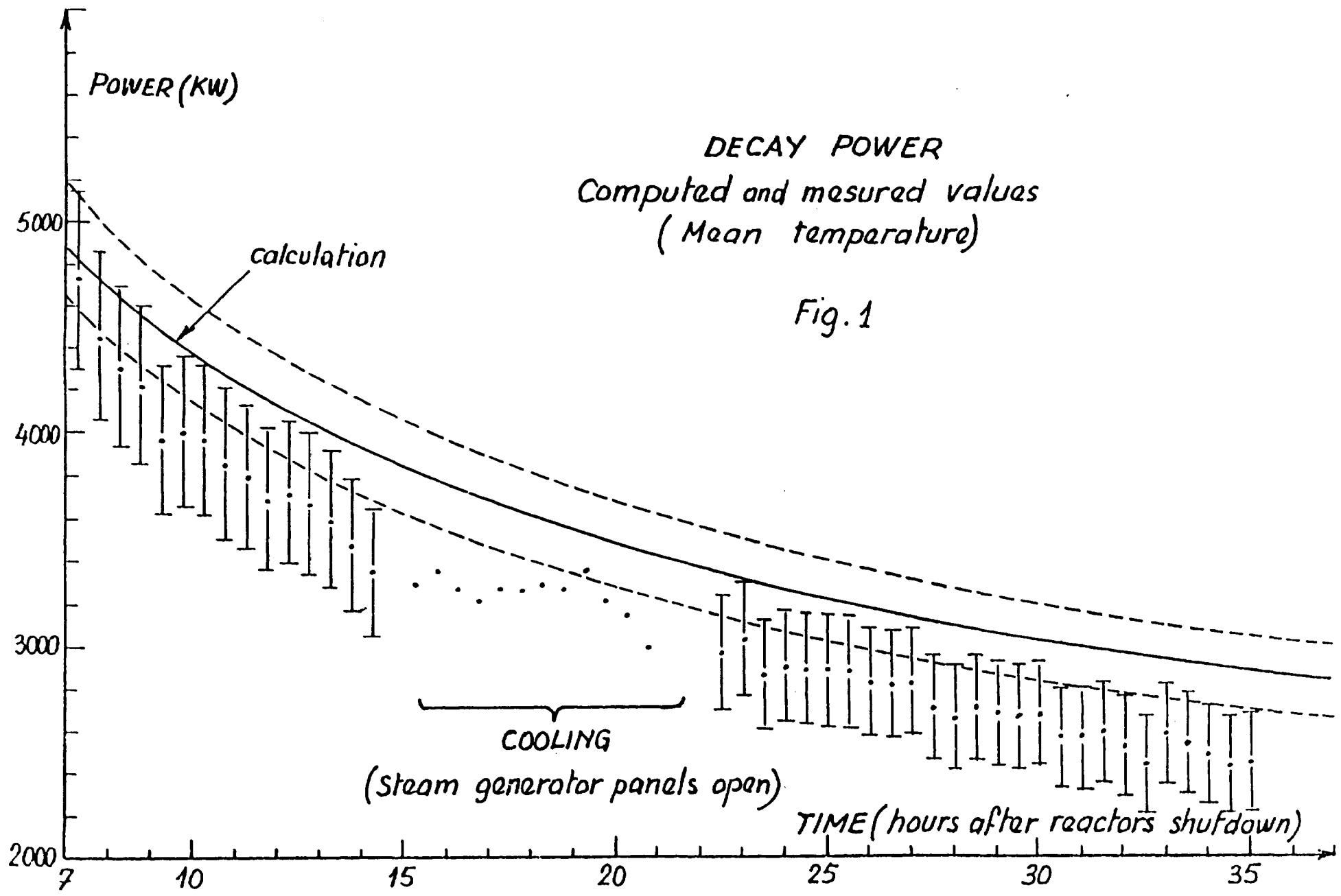
The computed values are coherent with the measured findings if allowance is made for uncertainty bounds on both the experimental results (about 10%) and the calculations (about 6%).

Because of the short time period considered, this integral experiment involves virtually only decay heat components 1 and 2 (fission products and $^{239}U + ^{239}Np$). Table 3 summarizes the calculated percentage contributions of each component.

This type of qualification is fully representative of fast breeder reactors. The basic code data and computation procedure as well as the specific reactor data would be similar or identical for SUPERPHENIX 1 or another French-designed fast breeder. Similar tests will be carried out on SUPERPHENIX.

TABLE 3

COMPUTED DECAY POWER PERCENTAGES FOR VARIOUS COMPONENTS					
TIME (hours)	FISSION PRODUCTS	138_U	Na	Steel	α
1	82.94	14.57	1.45	0.81	0.22
2	81.60	15.43	1.72	0.97	0.27
3	80.15	16.65	1.85	1.05	0.30
4	79.00	17.62	1.90	1.09	0.33
5	77.86	18.67	1.95	1.12	0.35
6	76.96	19.53	1.97	1.16	0.37
7	76.28	20.14	1.97	1.19	0.39
8	75.62	20.77	1.96	1.23	0.40
9	75.11	21.27	1.94	1.26	0.42
10	74.69	21.69	1.91	1.28	0.43
12	73.95	22.43	1.85	1.31	0.46
14	73.37	23.03	1.77	1.33	0.48
16	72.97	23.47	1.69	1.36	0.50
18	72.63	23.82	1.60	1.40	0.52
20	72.41	24.10	1.51	1.43	0.54
22	72.22	24.32	1.42	1.46	0.56
24	72.12	24.47	1.34	1.48	0.58
26	72.03	24.58	1.26	1.51	0.60
28	72.00	24.65	1.19	1.53	0.61
30	72.16	24.54	1.10	1.55	0.63
36	72.49	24.22	0.88	1.62	0.67
42	72.89	23.91	0.71	1.68	0.71
48	73.22	23.59	0.57	1.75	0.75
54	73.60	23.29	0.46	1.83	0.80
60	74.20	22.67	0.37	1.89	0.84
72	75.50	21.67	0.23	2.01	0.91

DECAY POWER
Computed and mesured values
(Mean temperature)

Fig. 1

POWER (KW)

calculation

COOLING
(Steam generator panels open)

TIME (hours after reactors shutdown)

5.1.2 - PHENIX Subassemblies

(a) γ Radiation Release

The energy released from PHENIX subassemblies by
γ radiation was measured during handling operations at the PHENIX
plant after cooling periods ranging from 2 to 6 months.

However, the nature of the measurements involved
in this type of experiment requires a substancial amount of calculation.

The actual γ emission spectrum is not directly
measured, only the γ spectrum observed outside the subassembly after
self-absorption, which in this case accounts for about 70% of the total
energy released as γ radiation. Moreover, the contribution of other
radiations must also be calculated.

The experimental findings are still in the process
of being interpreted but the following remarks can be made on the basis
of the results available to date.

- The discrepancy between calculated and measured
  total γ energy release is on the order of 20%.

- The deviations on the γ energy spectrum are
  40% at the most (low-energy values).

- Discrepancies of 10% to 30% are observed for
  measurements of γ rays specific to nuclides
  such as ^{95}Zr-Nb, ^{140}Ba-La, ^{106}Ru-Rh, ^{134}Cs,
  ^{137}Cs, ^{144}Ce-Pr, etc ..

(b) Total Energy Release

Work is currently in progress to measure the total
energy released from irradiated subassemblies using a calorimetric
method in which on-line measurements are made during subassembly transfer
from the reactor storage area to the transport casks.

This method should provide data valid within 10% for measurement times on the order of half an hour.

Both of these Phenix subassembly measurement methods are applied for implementation of a decay power comparator for fast breeder reactors used for on-line measurements.at subassembly handling facilities.

5.2 - Specific Measurements

The PROTEE computer code was also qualified by routine comparison with specific experimental values measured for each decay heat component either by the CEA or elsewhere.

5.2.1 - Component No 1 : Fission Product Energy

Of the numerous comparisons between computed and experimental results, some of the most significant are discussed below.

(a) Decay power calorimetric terms referred to the "elementary fission" for cooling times ranging from 0 to 1 day. The calculated curve is coherent with the experimental values, and its integral is about 15% higher for this time range. The same is true for comparisonswith the data given in reference [2], [3] and [4].

(b) Certain compilations or best-estimate curves based on theoretical and experimental results, e.g. the values given in reference [5], [6] and[7]. The table below summarizes the comparison between the PROTEE computed values and the ANS data given in reference [6]. Similar comparisons with the data in references [5] and [7]show that the PROTEE and ANS values are systematically 5 to 10% higher.

SUPERPHENIX 1 FISSION PRODUCT DECAY POWER IRRADIATION TIME : 616 DAYS NOMINAL CALCULATED VALUES (NO UNCERTAINTY BOUNDS)	
SHUTDOWN TIME	$\dfrac{\text{PROTEE} - \text{ANS}}{\text{ANS}}$
1 sec.	+ 12%
10 "	+ 6%
10^2 "	- 10%
10^3 "	+ 2.7%
10^4 "	+ 3.7%
1 jour	- 1.7%
12 "	- 1%
4 mois	+ 4%
8 "	- 2%
1 an	- 0.7%
2 "	- 12%
3.2 "	- 17%
6 "	- 12%

(c) Fission product concentrations measured during re-processing of spent fuel from the RAPSODIE and PHENIX reactors, after cooling times ranging from 5 months to 2 years. The discrepancies observed did not exceed 20%.

5.2.2 - <u>Component No 2</u> : ^{239}U and ^{239}Np $\beta + \gamma$ energy

The essential parameter here, i.e. the ^{238}U capture ratio is computed using the cross sections from the CARNAVAL IV set qualified by the CEA studies in the MASURCA facility, and the CEA spent fuel analysis program in RAPSODIE (Fortissimo and PHENIX).

On the basis of these comparisons the overall uncertainty (i.e. basic values + ^{238}U capture ratio) was estimated at $\pm$ 5%.

5.2.3 - Component No 3 : Heavy Nuclide α Energy

The α disintegration spectra for heavy nuclides (primarily ^{242}Cm, ^{244}Cm, ^{238}Pu) with energy levels on the order of 5 MeV were obtained from the literature and from the experiment described in reference [8]. The CARNAVAL IV cross sections were used. These values and the method used to compute the nuclide concentrations were qualified by the CEA spent fuel analysis program in RAPSODIE (Fortissimo) and PHENIX.

The uncertainty on the α emission energy is currently estimated to be $\pm$ 20%.

Similar measurement programs are scheduled for spent fuel from SUPERPHENIX 1.

5.2.4 - Component No 4 : Steel Structure Activation Energy

Based on measurements of stell irradiated in RAPSODIE the uncertainty on computed values of steel activation energy can be estimated at $\pm$ 100%.

As mentioned in paragraph 4.2.4, most of this uncertainty is attributable ^{60}Co, and a major experimental program has been undertaken with regard to PHENIX structural materials. The results, which should be available at the end of 1981, will appreciably reduce this margin. The experiment uses γ spectrometry at different points inside a PHENIX subassembly wrapper tube (axial blankets and fissile column). The initial ^{59}Co content of the steel is determined by fine analysis of the reference lots.

The absolute value of the ^{60}Co activity in the irradiated steel is determined using a Ge-Li detector by identifying the 1.17 MeV and 1.33 MeV lines in specific samples from the hexagonal wrapper.

5.2.5 - Component No 5 : Sodium Activation Energy

The ^{24}Na and ^{22}Na decay chains and energies are well known. The uncertainty on computed results is estimated to be ± 20% on the basis of comparisons with measured values, and is mainly due to the ^{23}Na reaction rate.

6 - <u>CONCLUSION</u>

The uncertainty on computed results was estimated for various decay power compounts by comparison with experimental findings. The table below summarizes the discrepancies for a French-designed fast breeder reactor.

<u>TABLE 5</u>

COOLING TIME RANGE	DECAY POWER COMPONENTS				
	1 FISSION PRODUCTS	2 ^{239}U & ^{139}Np	3 HEAVY NUCLIDE & RELEASE	4 STEEL STRUCTURES	5 ^{24}Na and ^{22}Na
Osec - 100sec	± 10%	± 5%	± 20%	± 100%	± 20%
100sec - 1year	± 5%	± 5%	± 20%	± 100%	± 20%
1year - 6years	± 10%	No contribution	± 20%	± 100%	NEGLIGIBLE CONTRIBUTION (^{22}Na ONLY)

The estimated uncertainty values only take account of basic nuclear data (cross sections, energy release, etc ..) or composition tolerances for the various media considered ; they are thus applicable to random variables.

The uncertainty on the total decay power is obtained by weighting the relative contribution of each component, and thus by combining the resulting uncertainties quadratically.

REFERENCES

(1) Mesures calorimétriques de la puissance résiduelle totale émise
par les produits de fission thermique de U235 ry Pu239
M. FICHE et al. Rapport DRE/SEN 022 - juin 1976.

(2) Decay Heat by Calorimetry, LA - UR - 76-2031, LOS ALAMOS SCIENTIFIC
LABORATORY (Sept. 1976)
J.L. YARNELL and P.J. BENDT.

(3) Fission product BETA and GAMMA energy release quaterly progress
report for July-September 1976, ORNL/NUREG/TM-65, Oak Ridge
National Laboratory (Dec. 1976)
J.K. DICKENS et al.

(4) ^{235}U fission product decay heat from 1 to 10^5, EPRI NP-180
(Feb. 1976)
S.J. FRIESENHAHN et al.

(5) Application of least-squares method to decay heat evaluation,
TC-796, Manfort Engineering Development Laboratory (Feb. 1977)
F. SCHMITTROTH and R.E. SCHENTER.

(6) Proposed ANS standard decay energy release rates following shut down
of uranium-fueled thermal reactors (OcT. 1971)
ANS - 5.1 - N 18.6.

(7) The most recently published evaluated nuclear data file
ENDF/B IV.

(8) Etude neutronique des noyaux lourds formés dans le cycle du
combustible des réacteurs nucléaires
DRE/SEN - Thèse de Docteur-Ingénieur de A. GIACOMETTI.