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Sensitivity and Uncertainty Analysis for Fission Product Decay Heat Calculations

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ABSTRACT

This paper presents the calculated uncertainty in decay heat due to the uncertainty in basic nuclear data given in the CEA86 Library. Uncertainties in summation calculation arise from several sources: fission product yields, half-lives and average decay energies. We take into account the correlation between basic data. The uncertainty analysis were obtained for thermal-neutron-induced fission of U235 and Pu239 in the case of burst fission and irradiation time. The calculated decay heat in this study is compared with experimental results and with new calculation using the JEF2 Library.

I. INTRODUCTION

The knowledge of the decay heat released in a fuel after reactor shutdown is necessary in connection with three fields of applications[1,3]:

- Removal of residual power as a function of the time elapsed since reactor shutdown for normal operation or emergency shutdown condition after a cooling time ranging from 0 to several days.
- Design of cooling systems, and, handling of irradiation fuel and its temporary storage at the reactor site after a cooling time ranging from a few hours to several months (or years).
- Fuel transport and reprocessing and waste packaging.

The decay heat can be obtained by two complementary methods, the first one is based upon the integral measurement of the total beta and gamma power. The second method, consists of cumulating the individual energies released by individual fission products; this is the summation calculation method. The integral measurements should be used to test the validity of the calculation approach which in turn could be applied to more complex situation in a reactor. Many codes were developed using the summation calculation[3]. To evaluate the fission product decay heat, we use the PEPIN code[5], which solves the radioactive filiation equations, by the analytical method, using a specific CEA library (DOP) containing 699 nuclides. The code was qualified by comparison with decay heat measurements and with other codes[4,5].

Several studies of decay heat uncertainties using the summation calculation have been made[7-10], and saveral sensitivity analysis have been made using the French data library[1,2,6]. In these studies the uncertainties in decay heat were calculated without taking into account the correlation between the basic data, because no correlation information was available. It implies that uncertainties were underestimated. However, Schmittroth and Schenter[8] considered a large correlated energy uncertainties for nuclides with an estimated Q values.

The object of this study is to calculate the uncertainties on decay heat associated with the PEPIN code through the variance/covariance analysis.

We take into account the correlations between the basic nuclear data from CEA86 library.

II. SUMMATION CALCULATION REVIEW

The energy released by the fission products at time t after a burst fission is given by the summation formula:

$$f(t) = \sum_{i=1}^{M} N_i(t) \cdot E_i \cdot \lambda_i \qquad (1)$$

where,

M: number of fission products in the library (M=699).

E_i: average beta (β) and gamma(γ) energy (MeV) emitted by the disintegration of nuclide (i).

λ_i: decay constant of nuclide i, $\left(\lambda_i = \frac{\log 2}{\text{half-life}} \quad (s^{-1})\right)$

$N_i(t)$: number of atoms of nuclide i at time t.

The concentrations $N_i(t)$ satisfy the BATEMAN equation:

$$\frac{\partial N_i(t)}{\partial t} = \sum_{j=1}^{i-1} \lambda_{ij} \cdot N_j(t) - \lambda_i \cdot N_i(t)$$

We can prove that:

$$N_i(t) = -\sum_{k=1}^{i} b_{ik} \cdot e^{-\lambda_k t} \qquad (2)$$

with,

$$b_{ik} = \frac{1}{\lambda_{ii} - \lambda_{kk}} \sum_{j=k}^{i-1} \lambda_{ij} \cdot b_{jk}$$

Where the summation is extending to all possible precursors of nuclide i in the same chain (mass A), *including the metastable ones, or in the (A+1) chain in* the case of delayed neutron emission,
and,

$$b_{ii} = Y_i - \sum_{k=1}^{i-1} b_{ik}$$

where Y_i is the independent yield of nuclide i, $\lambda_{ij} = b_{ij}\lambda_j$, and b_{ij} is the branching ratio from j towards i.

The decay heat at cooling time tc following an irradiation at a constant fission rate of one fission per second can be calculated by the integration formula:

$$F(T_{ir}, tc) = \int_{tc}^{T_{ir}+tc} f(t)\, dt \quad \text{Mev/s per fission/s} \qquad (3)$$

where,

Tir : irradiation time

f : decay heat function for one burst fission.

The effect of neutron capture is omitted in formula (3) because it is not important for cooling time lower then 10^4 s. For longer cooling time nuclides like ^{133}Cs and ^{147}Pm should be considred[1].

If the function f is known at the boundaries of small time intervals, the integration curve performed by assuming a linear variation of f in each time interval[1], in a log x log scale:

$$F(T_{ir}, tc) = \sum_{k=k_1}^{k_2} \frac{(f \cdot t)_{k+1} - (f \cdot t)_k}{\alpha_{k+1}} \qquad (4)$$

with,

$$\alpha_k = \frac{\log\left(\frac{f_{k+1}}{f_k}\right)}{\log\left(\frac{t_{k+1}}{t_k}\right)}$$

$$t_{k1} = tc \quad , \quad t_{k2} = T_{ir} + tc$$

III. UNCERTAINTY ANALYSIS

A. Principle of Uncertainty Calculation

Let $P_{l=1,..k} = P_l(X)$ be functions of the variable $X(x_1, x_2, .., x_n)$ where the x_i values are correlated or constrained.

Let E_{P_l} be the uncertainties in P_l which result from the propagation of error in X, $E_{P_l} = (var(P_l))^{1/2}$ where $var(P_l)$ is the variance of P_l.

We assume that a Taylor-series expansion is valid for P_l, at the first order:

$$P_l = \bar{P}_l + \sum_{i=1}^{n} s_{li} (x_i - \bar{x}_i) \qquad (5)$$

with, $s_{li} = \frac{\partial P_l}{\partial x_i}$

the variance of P_l is

$$var(P_l) = E(P_l - \bar{P}_l) = \sum_{ij} s_{li} s_{lj} (x_i - \bar{x}_i)(x_j - \bar{x}_j) \qquad (6)$$

and the covariance of P_{l_i} and P_{l_j} is,

$$cov(P_{l_i}, P_{l_j}) = E(P_{l_i} - \bar{P}_{l_i})(P_{l_j} - \bar{P}_{l_j}) \qquad (7)$$

It is straightforward to present the equations 6 and 7 in a matrix form[11]:

$$COVP = S_{P/X} \cdot COVX \cdot S^t_{P/X} \qquad (8)$$

Where, $S_{P/X}$ is the sensitivity matrix of P, dimension of matrix $S_{P/X}$ ($dim S_{P/X} = (k,n)$), and $S^t_{P/X}$ its transpose, COVX is the covariance matrix of X, dim(COVX)=(n,n), and COVP is the covariance matrix of P, is symmetric and dim(COVP)=(k,k), the variance of P are the diagonal elements, and the covariance are the non diagonal elements.

Furthermore, if P_l are functions of independent variables, $P_{l=1,..k} = P_l(X, Y, Z)$, $X(x_1, x_2, .., x_n)$, $Y(y_1, y_2, .., y_n)$, $Z(z_1, z_2, .., z_n)$, where X, Y and Z are independent, but internally correlated or constrained, then the covariance matrix of P is ,

$$COVP = COVP_{/X} + COVP_{/Y} + COVP_{/Z} \qquad (9)$$

In our case, P can be f(t), F(Tir,tc), or Y(Z,A), independent yield defined in paragraph III-B, and X, Y, Z can be the independent yield, half-livfe, energies, f(t),..etc..

Contribution to the total error of decay heat comes from uncertainties in three terms: fission yield, half-lives, and average beta and gamma energy.

The fission yields, half-lives and energies are supposed independent so that the uncertainty on decay heat is given by Eq. 9 (the uncertainties are the square roots of the diagonal elements of the covarince matrix).

B- Uncertainties in Decay Heat Due to Yield Uncertainties

The independent yields are calculate with the Wahl's Zp empirical model[12]. This model derived from experimental data, relates the systematic of nuclear charge distribution in fission to parameters for simple mathematical functions. For a given mass chain of mass number A, the independent yield of an isobar of atomic number Z is given by a gaussian charge dispersion formula,

$$Y(Z,A)=\frac{Y_c(A)\cdot Kp(Z,A)_f\, NF_f(A)}{\sqrt{\pi\cdot c}}\int_{Z-1/2}^{Z+1/2}\exp^{-\frac{(Z-Z_p)^2}{c}}dZ \qquad (10)$$

Yc(A): chain yield for the mass number A, is assume to be the cumulative yield for the last isobar in the mass chain of mass number A.

$Kp(Z,A)_f$: this factor takes into account the odd-even effects in proton (EOZ) and neutron pairing (EON).

proton	neutron	$Kp(Z,A)_f$
even	even	EOZ * EON
even	odd	EOZ / EON
odd	even	EON / EOZ
odd	odd	1/EOZ * EON

$Z_p(A)$: the most probable charge on mass number A.

c: parameter related to gaussian width σ, $c=2\sigma^2+1/6$

$NF_f(A)$: normalisation factor is used to achieve the Eq. 11 for each A.

$$\sum_{Zmin}^{Zmax} Y(Z,A)=Y_c(A) \qquad (11)$$

$NF_f(A)$ is equal to 1 when $Kp(Z,A)_f$ equal to 1. $Kp(Z,A)_f$ destroy the inherent normalisation properties of Gaussian distribution.

So the independent yield Y is a function of four parameters, Yc, Zp, Kp and σ, the corresponding covariance matrix COVY is given by the Eq. 8:

$$COVY = S\cdot COVW\cdot S^t \qquad (12)$$

S : sensitivity matrix of the independent yield.

$$S=\begin{bmatrix}\overbrace{\frac{dY_i}{dYc_j}}^{100} & \overbrace{\frac{dY_i}{dZp_j}}^{100} & \overbrace{\frac{dY_i}{dKp_j}}^{4} & \overbrace{\frac{dY_i}{d\sigma}}^{1}\end{bmatrix}_{699}\quad \overbrace{\phantom{\frac{dY_i}{dYc_j}\frac{dY_i}{dZp_j}\frac{dY_i}{dKp_j}\frac{dY_i}{d\sigma}}}^{205}$$

COVW : the covariance matrix corresponding to the uncertainties in the parameters that specify the fission products charge distribution, these parameters are independent, so one can express the overall covariance matrix, in terms of a set of independent submatrices,

$$COVW=\begin{bmatrix} COVYc & & & 0 \\ & COVZp & & \\ & & COVKp & \\ 0 & & & COV\sigma \end{bmatrix}$$

dim(COVW)=(205,205).

- COVYc : covariance matrix for the chain yields. The Yc values and their errors are from RIDER's compilation[14], but as we have no information about the correlation between uncertainties, we assume a full correlation. Our library contains 100 chains so dim(COVYc)=(100,100).

- COVZp : covariance matrix corresponding to Zp. The values of Zp and their errors are from ref. 12 and 15 We suppose a fully correlation between errors.

- COVKp : covariance matrix corresponding to Kp, obtained by the Eq. 8. EON, and EOZ and their errors are from references 13 and 15. The two parameters are independent, so that, their errors are uncorrelated. dim(COVKp)=(4 , 4).

- COVσ = var σ, from reference 12.

The covariance matrix COVY doesn't consider the constraints on fission yields (Eq. 11), who tend to reduce the errors in independent yields. We can prove that, the new covariance matrix of independent yields COVY ' is,

$$COVY' = (COVY^{-1} + G^t \cdot COVYc^{-1} \cdot G)^{-1} \qquad (13)$$

where the elements of G are given by,

$$\frac{\partial Yc(A)_j}{\partial Y(A_i,Z_i)} = \delta_{ij}$$

δ is the Kronecker symbol

If the errors in chain yields are uncorrelated and those in Y are uncorrelated too, the elements of the covariance matrix COVY ' are given as follow:

diagonal elements,

$$\mu_{ii} = \sigma_i^2 \left(1 - \frac{\sigma_i^2}{\sigma^2 + \sum_{j=1}^{n} \sigma_j^2} \right) \qquad (14)$$

and non diagonal elements,

$$\mu_{ij} = \frac{-\sigma_i^2 \sigma_j^2}{\sigma^2 + \sum_{j=1}^{n} \sigma_j^2} \qquad (15)$$

where σ^2_i is the independent yield variance, and σ^2 is the chain yield variance.

B-1 Uncertainty in decay heat.

The covariance matrix of decay heat corresponding to independent yields is given by Eq. 8,

$$COVf_{/Y} = S_{f/Y} \cdot COVY \cdot S^t_{f/Y} \qquad (16)$$

the dimension of $COVf_{/Y}$ is the number of cooling times studied.

the sensitivity elements are given by:

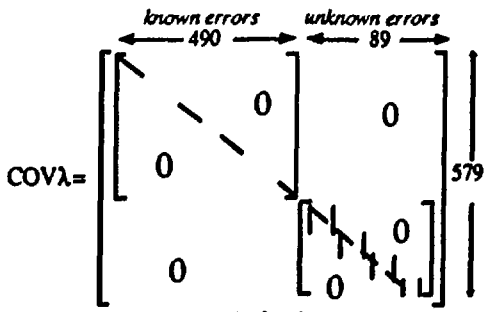
$$S_{Y_m}(t_j) = \sum_{i=m}^{M} E_i \cdot \lambda_i \cdot \frac{\partial N_i(t_j)}{\partial Y_m} \qquad (17)$$

C- Uncertainties in Decay Heat Due to half-life uncertainties

The uncertainties in the half-lives have been compiled by Blachot[16]. Almost all of the half-life errors are known, the unknown errors are from nuclides which have a very short or long half-life. To complete the list of errors, we assigned half-life errors as a function of the half-life itself. The half-lives were grouped according to time range and the average of the known errors was determined in each range. We give to an unknown error the average error of the corresponding time interval. In each time interval, unknown errors are assumed to be fully correlated and the known errors uncorrelated.

The unknown errors are correlated because they are obtained by the same method. Some known errors could be correlated, this sort of detail involves a large effort and is not necessary due to the small contribution of half-life errors to the total decay heat uncertainty (fig. 2).

So the covariance matrix for half-life errors has the following form:



The covariance matrix for decay heat corresponding to half-life is:

$$COVf_{\lambda} = S_{f/\lambda} COV\lambda \cdot S^t_{f/\lambda} \qquad (18)$$

The sensitivity elements matrix are derived as follows:

$$S_{\lambda_m}(t_j) = -\sum_{i=m}^{M} E_i \cdot \left(\lambda_i \cdot \frac{\partial N_i(t_j)}{\partial \lambda_i} + \delta_{im} \cdot N_i(t_j) \right) \qquad (19)$$

$\delta_{im} = 1$ for $i = m$, $\delta_{im} = 0$ for $i \neq m$

D- Decay Energies Uncertainties

An obvious source of error in summation calculations is due to uncertainties in the decay energies for each nuclide. For the CEA Library , it is useful to divide the nuclides into two classes: the well known nuclides for which beta and gamma spectra are well known experimentally, and the unknown nuclides for which beta and gamma or both spectra are unknown.

For known nuclides the average gamma-ray, beta-particle, and neutrino energies are calculated as following:

$$\bar{E}_\beta = \sum_i \varepsilon_{\beta_i} \cdot I_{\beta_i} \qquad (20)$$

$$\bar{E}_\gamma = \eta \sum_i \varepsilon_{\gamma_i} \cdot I_{\gamma_i} \qquad (21)$$

and,

$$\bar{E}_\nu = \sum_i \varepsilon_{\nu_i} \cdot I_{\nu_i} \qquad (22)$$

Where gamma-ray and beta particle intensities are denoted by $I_{\gamma i}$ and $I_{\beta i}$, ε_i is spectre energy. Internal conversion energy is added to beta energy, and its uncertainty is negligible.

The beta-particle intensities are constrained by:

$$\Sigma I_{\beta i} = 1 \qquad (23)$$

η is the normalisation constant to achieve the constraints,

$$\bar{E}_\gamma + \bar{E}_\beta + \bar{E}_\nu = Q$$

$$\bar{E}_T = Q \cdot \bar{E}_\nu \qquad (24)$$

with $\bar{E}_T = \bar{E}_\gamma + \bar{E}_\beta$

The uncertainties in average total energy are calculated by assuming that the β and γ components are non correlated.

$$\Delta\bar{E}_T = \left(\Delta\bar{E}_\beta^{\,2} + \Delta\bar{E}_\gamma^{\,2}\right)^{1/2} \qquad (25)$$

There is generally a strong negative correlation between the two components, due to the constraint with Q (Eq. 24). This negative correlation should tend to reduce the uncertainties in total decay energy, so that this equation can be considered to overestimate the uncertainties.

The variance of β and γ energy is given as:

$$\mathrm{var}\bar{E}_\gamma = \mathrm{var}(\bar{E}_\gamma)_{/\eta} + \mathrm{var}(\bar{E}_\gamma)_{/\varepsilon_\gamma} + \mathrm{var}(\bar{E}_\gamma)_{/I_\gamma} \qquad (26)$$

$$\Delta\bar{E}_\beta^{\,2} = \mathrm{var}\bar{E}_\beta = \mathrm{var}(\bar{E}_\beta)_{/\varepsilon_\beta} + \mathrm{var}(\bar{E}_\beta)_{/I_\beta} \qquad (27)$$

the variance of beta-ray with respect to beta intensities is reduced by the constraint, (Eq. 23):

$$\mathrm{var}(\bar{E}_\beta)_{/I_\beta} = \sum_i \varepsilon_{\beta_i}^2 \mathrm{var}(I_{\beta_i}) - \frac{1}{\sum_i \mathrm{var}(I_{\beta_i})}\left[\sum_i \varepsilon_{\beta_i}\mathrm{var}(I_{\beta_i})\right]^2 \qquad (28)$$

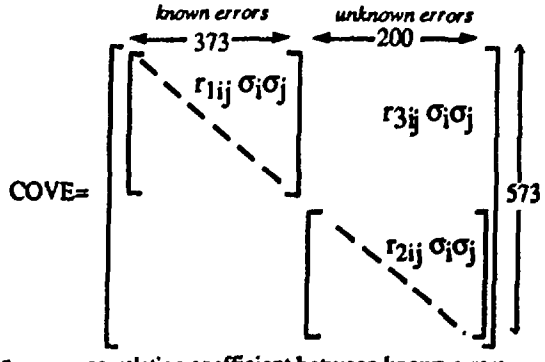
This equation was obtained by replacing in Eq. 14 σ^2 by 0 (the variance of a constant is 0) and by Eq. 8.

In general, unknown nuclides have very short half-life; that is the reason why measurements don't exist. We estimate their beta and gamma energies from the known nuclide energies. We plot beta and gamma energies as a function of Q and we calculate the straight line regression associated. The unknown energies are then obtained, knowing their reaction heat Q.

For unknown errors, the following rules have been applied, so the unknown errors are correlated.

Error in energy β = 15%
Error in energy γ = 30%

The covariance matrix for average total energy is:



r_{1ij} correlation coefficient between known errors
r_{2ij} correlation coefficient between unknown errors
r_{3ij} correlation coefficient between known and unknown errors

As we have seen previously, there is a possible correlation between the errors for known nuclides. The problem is that there is no information about these correlations.

As the same value is assigned to the errors for the *unknown nuclides, these errors are correlated.*

The library gives no information at all to estimate all these correlation so, we study the two extreme cases.

In the first case, we suppose that there is no correlation between errors; in the second one, the correlation is supposed to be maximal.

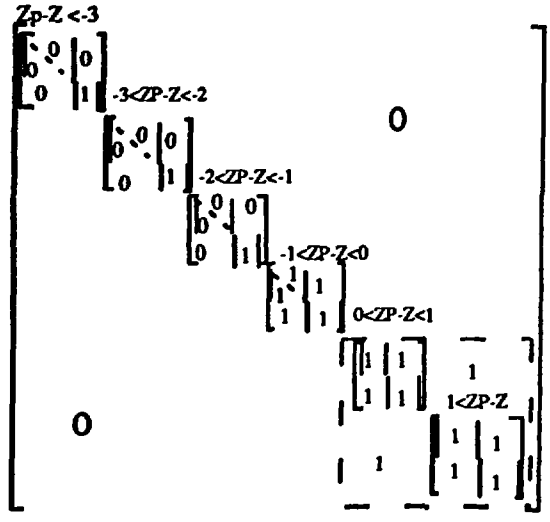
We can notice a great difference between these two cases (fig. 1). That means the estimation of this correlation need to be precise if we want a good error estimatation.

Another attempt was made to evaluate unknown uncertainties and correlation coefficients, by the following procedures:

The uncertainty in decay energies depends on the nuclide distance from stability line[7]. The more the nuclides are far from Zp the more they are unstable, and their decay energy uncertainties increase (see the table below). Therefore, we assumed that the correlation coefficients increase in the same way. We classified average beta and gamma energy uncertainties in bins as a function of the distance in charge units from Zp. In each bin unknown errors were determined by the average value of the bin, and assumed to be fully correlated. The correlation coefficients between errors were evaluated according to the (Zp-Z) intervals.

(Zp-Z) intervals	number of Known nuclides	number of unknown nuclides	average uncertainty %	adopted unknown uncertainty %
-3 >	52	7	3,47	4
-3 , -2	84	14	5,21	6
-2 , -1	105	12	8,11	8,5
-1 , 0	75	46	11,5	12
0 , 1	41	51	12,8	15
>1	16	70	14,1	18

correlation matrix for energy



From the calculation presented in fig.1 and 2 (thermal burst fission of U235), some comments are given:

- The uncertainties associated with the half-lives play only a very small role.
- The uncertainties associated with the yields play a relatively minor role.
- The uncertainties associated with the decay energies are the determining for all cooling time, due to the linear dependency of decay heat in the average total energy (see Eq. 1) and the lack of data mainly for nuclides far from the line of beta stability.

E.Uncertainty After an Irradiation Time

The uncertainties in decay heat following an irradiation at a constant fission rate of one fission per second is given by Eq. 8:

$$COVF = S_{F/f} COVf \cdot S^{t}_{F/f}$$

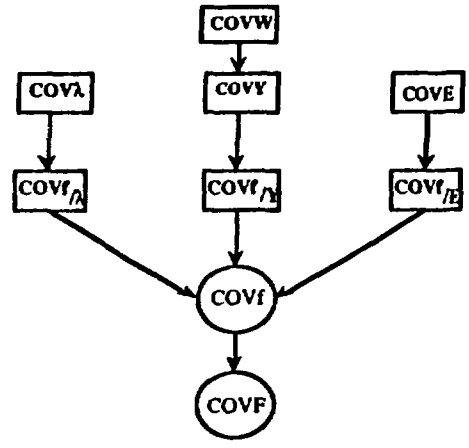
where,

$$COVf = COVf_{E} + COVf_{\gamma} + COVf_{\lambda}$$

The sensitivity matrix element are given by deriving the Eq. 4 by f(t).

For a safety analysis, we retain the most pessimistic case (maximal correlation between energy uncertainty).

Finaly the flow diagram of uncertainties analysis in this study for fission product decay heat calculation is presented in the following figure.



IV DECAY HEAT COMPARISONS

The uncertainty analysis for decay heat summation calculation gives a certain degree of confidence to such calculation without recourse to experimental verification when it is not available. We compare our results of decay heat calculation based on the CEA86 Library with calculation results based on JEF2 Library, and, direct experimental results, from Dickens et al. at Oak Ridge National Laboratory[17], and Akiyama et al.[18] in Japan. Results of both experiments are presented in Fig. 4, 5, 6 with calculation values of this study, in the cases of thermal burst fission of U235 and Pu239 and thermal infinite irradiation of U235.

Our calculated values are in agreement with those based on JEF2 Library and with the experimental ones.

V. DISCUSSION AND CONCLUSION

Evaluated decay heat uncertainties were obtained for thermal-neutron-induced fission of U235 and Pu239 as function of cooling time and irradiation time (fig. 3 and fig. 4). The computer programmes which have been developed can be applied to any irradiation time and cooling time condition, and for the others fissioning systems: thermal-neutron-induced fission of U233 and Pu241, and fast-neutron-induced fission of U235, U238, Pu239, Th232, U233, Pu240, Pu241 and Pu242.

Theoretically, the uncertainties on decay heat in the reactor case are the twelve fissioning systems combination. These systems are correlated by the fact that we use the same energy and half-life data. We didn't take into account this correlation in this study, because it has been done for a given system. The most important fissile isotopes in thermal reactor are U235, Pu239 and incidentally U238. At the beginning of the UOX fuel evolution the predominant fission is U235, but, as the fuel evolves, the fission of Pu239 is not negligible.

To be sure to have a good uncertainty of decay heat in the power reactor case, whatever the nature of the fuel is, UOX or MOX, we have chosen the maximum uncertainties from U235 and Pu239 fissioning systems. Since the uncertainty curves of these two fissile nuclides are very close (fig. 3), our results are somehow maximised. The maximised process can guarantee the independence of our calculation whatever the evolution of the fuel composition.

In our future study, the neutron capture effect may be considered for cooling time $t_c > 10^4$ s, especially in connection with fuel assembly transport and storage problems.

The main sources of uncertainties in decay heat summation calculations are due to decay energy uncertainty propagation. So it is necessary to emphasize the study of energy covariance matrix. Some informations related to the direct determination of average beta energies[19] are available so it is possible to improve the energy covariance matrix.

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fig 1: uncertainties in decay heat due to energy uncertainties U235 (thermal) burst exposure

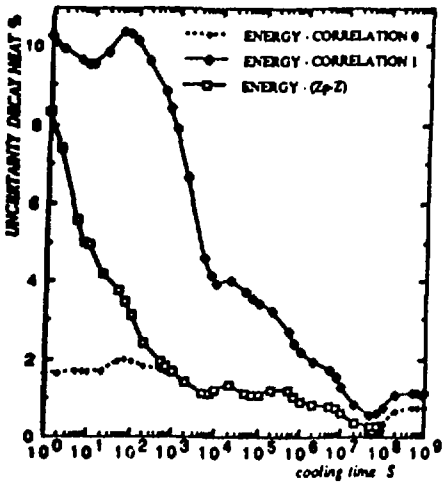


fig 2: uncertainties in decay heat due to basic data uncertainties. U235 (thermal) burst exposure

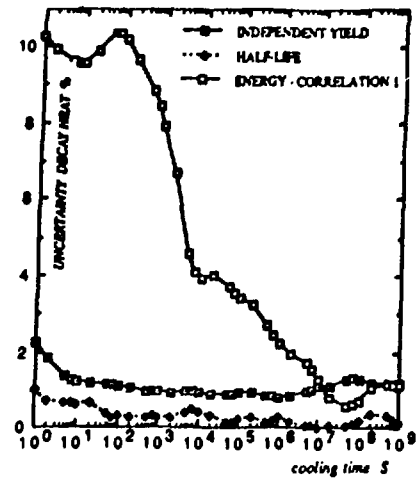


fig 3: uncertainties in decay heat due to basic data uncertainties. thermal U235 and Pu239 3SEWA Burnup

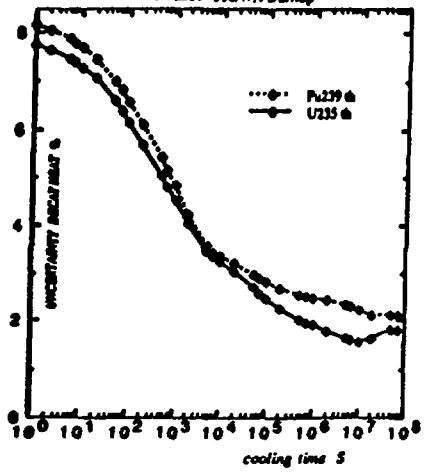


fig 4: U235 (thermal.) Decay Heat infinite irradiation

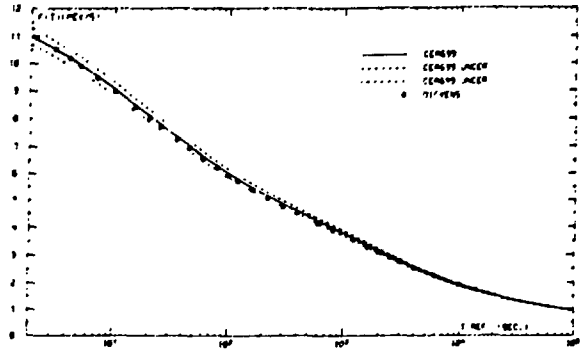


fig 5: U235 (thermal) Decay Heat Burst Fission

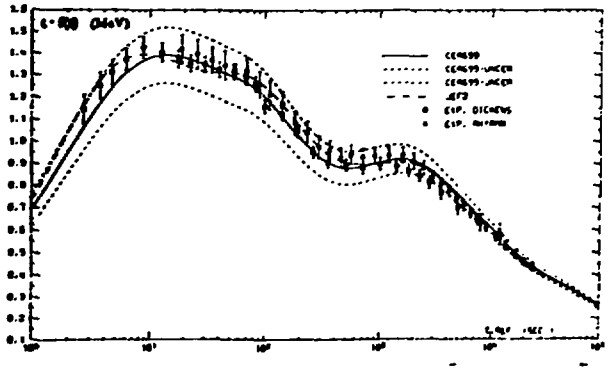
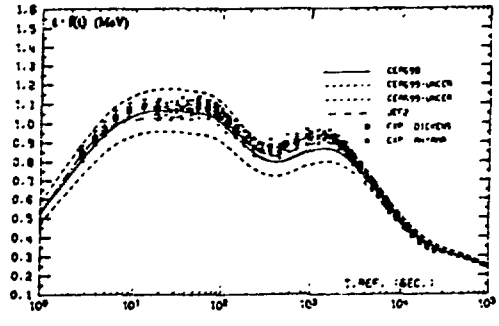


fig 6: Pu235 (thermal) Decay Heat Burst Fission



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