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# FUEL HISTORY DATA APPLIED TO REPROCESSING PLANT INPUT INSPECTION

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## ABSTRACT

The application of simplified burn up calculations with a limited amount of historical data on the spent fuel and its irradiation make it possible to predict all the final compositions of the spent fuel with a very good approximation.

Combining these values with destructive or non-destructive measurement results for fuel samples on entry into the reprocessing facility provides a large number of possibilities for checking the accuracy of the results.

Among the application presented, the "gravimetric balance" method of determining the plant input uranium and plutonium mass was the subject of considerable investigation during the first water

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reactor fuel reprocessing campaigns at La Hague. The accuracy of the results and their high degree of confidence based on the numerous cross-checks demonstrate the effectiveness of the methods, which also make it possible to reduce the number of complex analyses (e.g. of neodymium) and therefore the cost of the input material balance determinations.

By permitting the interpretation of indirect measurement results (such as the gamma emission of certain fission or activation products in terms of specific burnup, cooling time or fuel mass) these methods provide non-destructive inspection solutions. Two major examples are the identification of spent fuel subassemblies and the assessment of the quantities of fissile materials in the hulls.

## INTRODUCTION

By definition, spent fuel is fuel which, after a time interval in a reactor, has undergone certain modifications in composition related to nuclear reactions, primarily capture and fission.

On arrival of the spent fuel at the reprocessing plant, two widely divergent attitudes may be adopted to determine its composition :
- Disregarding its origin, it may be considered that the properties of the incoming product are largely unknown, and that it is necessary to measure all the parameters required to define the material balance and, more generally, the operations related to fuel reprocessing.

- Conversely, it may be assumed that the fuel is perfectly identified, and that all its properties may be determined by calculation from its initial composition and irradiation history.

The first attitude has the advantage of simplifying the problem by completely isolating the reprocessing operation from the reactors. It would, however, be extremely difficult to apply in practice because of the number and complexity of the measurements required. The second, if strictly applied, would be unrealistic for two fundamental reasons : the insufficient accuracy of calculated estimates for certain compositions, and the possbility - however remote - of an error in identification.

In practice, an intermediate method is required between these two extremes. The solution currently adopted at La Hague is to measure the essential parameters (e.g. the Pu/U ratio and the isotopic abundance of these two elements), and to take account of the fuel history to provide cross-checks and to determine the secondary parameters (e.g. abundance or activity of fission products or transplutonium elements).

The studies presented here, which constitute the basis for the current application of calculated relations between various parameters, tend also to show that it will gradually be possible to take greater account of the fuel irradiation history and thereby either reduce the number of required measurements or provide further valuable data for the operation and monitoring of the plant.

After a theoretical presentation based on experimental confirmations, three areas of application will be examined : determination of the input material balance, non-destructive identification of fuel, and determination of supplementary data required for monitoring reprocessing plant operation.

## 1 - OBJECTIVES

### 1.1 Nuclear Safeguards Inspection

The reprocessing plant input balance is one of the key points in the fuel cycle for nuclear material accounting. The last preceding measurements are normally made during the fuel fabrication process, while a calculated estimate of the amount of residual Pu and U is made on removal from the reactor.

Consideration of the fuel history, i.e. data on its initial composition and on the reactor irradiation conditions together with its final composition as determined by suitable calcultations, presents a number of advantages for nuclear material control.

- Cross-checks between the estimated material amounts on discharge from the reactor and the amounts measured on arrival at the reprocessing plant;
- Verification of the accuracy of results obtained by different analysis techniques.
- Reduced cost and response time through the development of new material balance determination techniques and the use of high-speed analysis methods.
- Fuel identification by non-destructive analysis on arrival at the plant or on departure from the reactor.
- Determination of the amount of nuclear material contained in the hulls.

For all these points, the interests of the nuclear safeguards inspection authority do not differ basically from those of the plant operator.

1.2 - Other Applications

Additional applications involve reprocessing plant operation and safety. Basically the inspection problems require thorough knowledge of the fuel to provide accurate material balances, to optimize the design or operation of the facility according to the fuel reprocessed to ensure adequate plant safety levels without excessive safety margins for criticality or biological shielding problems, and to permit precise waste management.

Non-destructive identification of fuel assemblies and proper interpretation of various measurements after dissolution, backed up by calculations which take account of the fuel history should provide full and reliable answers to all these identification problems.

Knowledge about the compositions makes it possible to monitor the fuel, to confirm the absence of fissile material in sub-products such as hulls or to determine the minute amounts which might be present, and to prevent accumulation hazards at certain process stations.

Such data may be obtained simply by interpreting measurement results using calculations based on the fuel history.

2 - METHODS

Fuel evolution under irradiation is governed by physical laws which are well known even though they cannot always be satisfactorily represented in calculation.

Very thorough neutron codes are used to prepare a detailed balance of the nuclear reactions occuring in the fuel and thus to determine the final post-irradiation compositions from the initial fuel fabrication values.

2.1 - Composition Accuracy as calculated with Reactor Codes

The estimated uncertainty on the calculated mean composition values for a fuel assembly from a high-power pressurized water reactor is shown in Table I for the methods currently available to the CEA. The final values depend on the specific burnup, which may be specified by the reactor operator on the basis of thermal balance measurements or determined by destructive or non-destructive measurements on the fuel.
The precision of the composition determination is obviously related to the degree of uncertainty about the burnup ; two examples are given in Table I.

2.2 - Simplified Method for Calculating the Fuel Composition

Reactor calculation methods are complex and relatively cumbersome in that they require a detailed representation of the entire configuration at each stage in the irradiation process. In reality, this complexity stems essentially from the fact that such codes are used to acquire other data than fuel composition modifications, particularly the evolution of reactivity and power distributions. It was therefore logical to try to simplify these methods for applications strictly limited to determining the final fuel compositions.

The CEA has thus developed a calculation procedure based on the following principles :

- Taking account of the major reactor and fuel characteristics to obtain accurate "quasi-point" calculations.
- Use of qualified cell-codes to determine the mean cross sections and their variation under irradiation for fuel assemblies or assembly zones. For water reactors, the APOLLO code is used (1).
- Complete evolution calculations using a dimensionless space code with allowance for an approximate power history. This code, EVOGENE, has been described elsewhere (2).

This method provides, for each fuel, composition tables as a function of specific burnup and cooling time together with formula sets which account for the interrelation of the major characteristic parameters of the final compositions.

This simplification - which is considerable in terms of the means required to obtain the desired values - essentially involves disregarding the influence on the cross sections of any perturbations liable to have affected the fuel during irradiation and due, for example, to its immediate environment, to control rod movements, to moderator temperature variations, etc.

The importance of these approximations is not of the same order of magnitude for all the parameters, as will be discussed in para. 2.3. The simplified method is qualified in two ways :

- from qualification of the cell code and the related neutron librairies resulting from reactor physics work ;

- overall comparisons between values calculated by the simplified method and experimental results.

The first point has already been discussed in the litterature (3) for the APOLLO code, and will not be repeated here.

Table II and III show some typical results illustrating the second point. Table II indicates the magnitude of the deviation between experimental and calculated values before adjustment of the formula set while Table III presents the mean deviations after adjustment for fuel from the Ardennes Nuclear Plant and from the STADE reactor, reprocessed at La Hague. The good coherence obtained, for the latter case, is of particular interest, since these results were not taken into account for the adjustment.

2.3 - Influence of Approximations on Various Parameters

In order to evaluate the importance of the approximations used with the simplified method for different parameters, compositions or relations among compositions which are of particular interest for reprocessing plant entry inspections, the authors have performed a differential effects analysis by artificially creating perturbations much greater than those liable to affect the fuel assemblies of a single reactor. The results of this study are presented in Table IV.

A number of conclusions may be drawn from these examples :

- The relationship between specific burnup and uranium isotopic composition variations is higly sensitive to the initial composition (example A), which is self-evident, and demonstrates the need for cross-checks with other independent determinations.

- Various relations are sensitive in varying degrees to the specific burnup ; however, this does not constitute a selection criterion since the specific burnup is one of the results, and the consequences of a poor initial estimate can easily be minimized by iteration.
- Spectrum perturbations under irradiation (such as example C, introduction of absorbant material , and example D, moderating ratio variations) are particularly sensitive with respect to relations involving the Pu/U ratio. The values indicated show that circumspection is required when using these relations, and especially that their practical interest must not be evaluated on the basis of their low sensitivity to possible errors on the initial fuel composition or the specific burnup.

## 3 - APPLICATION TO PLANT ENTRY BALANCES

The conventional method of determining the uranium gravimetric balance on entry into the reprocessing plant requires a long and costly analysis in which the most difficult phase involves fission neodymium determination by mass spectrometry.

With regard to PWR fuel at La Hague, work is in progress to develop methods which will lead to the gradual replacement of much of the neodymium measurement process by determinations of other parameters. During the most recent campaigns, for example, fission mass calculations were made from the following data :

- the integrated irradiation (data supplied by reactor operator)

- the Pu/U ratio determined by laboratoy analysis at the facility
- $^{235}U$ depletion, based on fabrication data and laboratory measurements
- the $^{134}Cs/^{137}Cs$ activity ratio.

Several possible ways of cross-checking the Pu/U ratio measurements were also examined.

3.1 - Specific Burnup Determination

3.1.1. - Required precision

A detailed study of the uncertainties affecting the uranium and plutonium balances - allowing for the extreme parameter values possible for spent PWR fuel (max. burnup 40 000 MWD/T) - shows that a 5 % tolerance on the specific burnup determination adds a maximum uncertainty factor of 0.2 % on the amounts of uranium and plutonium. If in the burnup determination the uncertainty resulting from systematic errors does not exceed 2 %, the effects of random errors may be minimized by measuring an adequate number of samples ; thus for a large fuel batch, e.g. a water reactor refueling batch (one-third of the core load), no more than 0.1 % of the final uncertainty on the U and Pu balances is attributable to the burnup determination. This is a reasonable objective, and it is important to point out that these values are attainable from the reactor operator's data, thus providing a cross-check solution.

3.1.2. - Reference method

After several years of research work on specific burnup measurement techniques, the CEA has adopted the neodymium method, in which the $^{148}Nd$ isotopic abundance is measured by mass spectrometry and compared with the amount of $^{238}U$ (4). This is a complex measurement,

and extensive means are required for routine application. The determination precision is 2 - 3 %, while the systematic error contribution, related in particularly to the absolute uncertainty regarding $^{148}Nd$ fission yields, is on the order of 1 - 2%. This technique is considered as the reference method and has been applied at La Hague to the 17 dissolutions of the first campaign (BWR fuel from Muhleberg) and to 15 of the 81 dissolutions of the second campaign (PWR fuel from stade).

3.1.3 - Cesium activity ratio method

The method comparing the activities of $^{134}Cs$ and $^{137}Cs$ to determine specific burnup has been discussed elsewhere (5).

CEA laboratory studies have been conducted on solutions of spent PWR fuel from the Ardennes Nuclear Plant with a wide range of specific burnup values and cooling times. Typical results are shown in Figure 1. The results are interpreted according to the simplified method described in paragraph 2.2.

For the two reprocessing campaigns mentioned, systematic gamma spectrometry measurements were performed on dissolver solution samples used for other analyses. Full interpretation of these results was not possible since some irradiation history data was lacking. Nevertheless, the direct use of the measured results as relative values showed excellent coherence with the neodymium values for the Muhleberg campaign, although the dispersion was much greater for the Stade campaign.

3.1.4. - Uranium and plutonium isotopic measurements

Routine specific burnup determinations were made on the basis of the uranium and plutonium isotopic abundance values measured for dissolver samples. This technique is based on the conclusions of theoretical and experimental studies already published (2) and outlined in paragraph 2.3 above.

The formula sets mentioned in para 2.2 were used to obtain specific burnup figures from measured isotopic composition variations ($^{235}U$ depletion, $^{236}U$ formation, Pu/U ratio). A statistical analysis of the results obtained for the Muhleberg and Stade campaigns is presented in Tables V and VI together with the values obtained for samples from the Ardennes plant.

It may be noted that this analysis, on a large number of results, provides for the random error estimate a standard deviation of less than 3 % per measurement - in most cases much less than this value. The Student test for revealing any systematic errors is generally negative despite the large number of measurements involved and thus the random error is small on the mean deviation between methods ; in the few instances where a positive value was obtained the systematic error estimate between methods was less than 1 %.

3.2 - Pu/U Ratio Determination

3.2.1. - Required precision

For water reactor uranium fuel, the Pu/U ratio measurement is the major

element of uncertainty in the plutonium balance obtained by the gravimetric method. The desired accuracy is better than 1 %, and a reasonable objective is the uncertainty on direct Pu/U ratio measurements by mass spectrometry using the double isotopic technique, i.e. approx. 0.5 %.

3.2.2. - Calculation precision

The precision levels which may be expected from "reactor" calculations, given the uncertainty about the specific burnup, were indicated in paragraph 2.1. It is clear that even if the specific burnup is assumed to be accurately known by means of all the methods described in paragraph 3.1, the precision of any individual calculated Pu/U ratio determination will be inadequate to attain the desired objective. However, for a fuel batch comprising a reactor refueling load or a significant fraction of a load, it is not unreasonable to expect adequate precision from the calculations defined in paragraph 2.2 and an accurate specific burnup determination. This is clearly indicated by the examples in Table VII.

3.2.3 - Isotopic measurement applications

A number of Pu/U ratio indicators other than specific burnup have been proposed by various authors. Here again, a systematic research program was undertaken on the results of the La Hague reprocessing campaigns, based on the conclusions of the studies already cited (2).

The results (Table VII) show that all these determinations are somewhat

less accurate than one based on the specific burnup. The deviations noted for the mean batch values could be reduced by more thorough qualification of the calculations for the relevant isotopes. The combinations of isotopic ratios may undoubtedly be improved by compensating for the effects of any perturbations, but this approach would run up against the accuracy limits of the analysis results.

### 3.3 - Uranuim and Plutonium Mass Determinations

The uranium and plutonium mass balance was determined for the same two reprocessing campaigns on the basis of the various specific burnup determinations previously discussed.

Table VIII contains the final results of the Stade campaign, and shows that a rather wide range of methods is available to limit the extent of the measurements required for application of the gravimetric method or to check the results by means of independant methods.

It may be noted that the standard deviation for a uranium mass determination is less than 0.1 %, and that no systematic deviation exceeding 0.05 % is attributable to differences in the specific burnup determination methods. For the plutonium mass balance it is also necessary to take account of the uncertainty on the Pu/U ratio.

## 4 - APPLICATION TO NON-DESTRUCTIVE IDENTIFICATION OF SPENT FUEL

It is another possible field of application for these methods of special interest in that it was inconceivable without the development of this research.

Some preliminary CEA studies were based on the experience acquired with gamma spectrometry measurements on water reactor fuel assemblies and on work involving interpertation of spent fuel composition measurements.

The identification measurement technique, procedures and inspection principles are covered in another article presented at this meeting (6). The following discussion concerns only the contribution provided by taking account of the fuel history for such controls.

4.1 - Specific Burnup Measurement

Research work designated two feasible methods for non-destructive measurements of specific burnup on water reactor fuel :

- $^{134}Cs/^{137}Cs$ activity ratio measurements.
- $^{137}Cs$ absolute abundance measurements.

Both techniques require a minimum of fuel history data for interpretation with a greater quantitative amount of data required for the first method. On completion of the preliminary work mentioned earlier, the proposed solution involved on-line data processing using the formula sets derived from the calculation method discussed in paragraph 2.2.

The major data items required for this purpose are :

- the initial fuel composition and geometry
- the type of reactor used for irradiation
- the approximate flux history and date of fuel, removal from the core.

The first two items are required for neutron cross section calculations, as in the other applications. The third is of special importance for Cesium activity ratio measurements because of the radioactive half-life of $^{134}Cs$ (2.1 years). Since this is a precise bit of reactor operating information, it is useful to determine the degree of accuracy with which it should be known. An examination of the various operating histories outlined in Table IX indicates that ideally the known data would comprise a monthly reactor power history and the specific burnup values announced at the end of each cycle. If this is not possible, it is sufficient in most cases to take account of the specific burnup figures and cycle beginning and end dates : the resulting approximation errors will not exceed 5 % on the actual specific burnup.

4.2 - Control Validity

Since it requires that fuel history data be taken into account, the specific burnup determination alone is not sufficient to permit fuel element identification. For this reason, the proposed solution also includes a measurement of cooling time based on gamma-emitting fission products of very different half-lives ($^{95}Zr$, $^{144}Ce$, $^{106}Pd$, etc.), together with an identification control involving comparison of the values determined for these parameters (i.e. specific burnup and cooling time) with the values indicated on the fuel log card. Two questions might be asked at this point :

- What is the quality of an identification based on these two parameters ?
- What happens if one of the uncontrolled data items is false ?

The diagram in Figure 2 provides a partial answer to these questions. It may be observed that the $^{134}Cs/^{137}Cs$ and $^{144}Ce/^{137}Cs$ activity ratios permit a precise discrimination of the two parameters concerned, and that an error on the initial enrichment value

may be detected by this control if it is of significant proportions.

## 5 - APPLICATION TO OTHER NUCLEAR CONTROLS AT THE REPROCESSING FACILITY

The gamma and neutron activities of the spent fuel batches may be used for a number of nuclear controls involving process operation, plant safety and the embodiment of nuclear material safeguards. In most cases, whether the desired values concern overall activity or the specific activity of a given isotope, data calculated on the basis of the fuel history is required to interpret the results. Once again, in order to ensure that these calculations can be used with relative simplicity and in conjunction with a limited set of historical data, it is essential to select the measured parameters carefully in view of the desired objective.

### 5.1 - Parameter Sensitivity to Data Input and Calculation Methods

As an example, the relationship among the various activities and the corresponding fuel amount were examined for their sensitivity to the degree of accuracy of the historical data and to the approximation inherent in the simplified calculation method (para. 2.2), assuming that all other problems involving measurement conditions and the interpretation of radiation propagation are resolved. The results are presented in Table X.

It is readily apparent that, for these purposes, the total neutron activity measurement is much more sensitive - and therefore more delicate in use - than gamma activity measurements of a long

half-life fission product. This, of course, is only an example.

5.2 - Gamma Spectrometry Inspection of Hulls

Among the many applications to nuclear controls, we have selected one more directly related to the enforcement of safeguards : a control of the amounts of fissile materials which may be retained as waste products with the cladding residue after dissolution. This process is described in another paper presented at this meeting (7) and once again the discussion here will be limited to the problem of taking the fuel history into account to interpret the results of this control.

The method adopted for water reactor fuel is to measure the absolute quantity of $^{144}Ce$ and then to calculate the amount of residual fuel. Figure 3 shows an example of the nomograms calculated to interpret the results, and Table X summarizes the influence of the major parameters on this relation. It may be observed here that, while calculation is necessary for proper use of the results, the approximations on the method or the data items have only minor effects, with the exception of the cooling time.

CONCLUSION

Allowance for the irradiation history of reprocessed fuel is capable of providing significant data for nuclear safeguards control at the reprocessing plant entry point. This procedure requires only a minimum of fuel history data, but can only furnish accurate and reliable data if the results are correctly interpreted using reactor physics calculation methods.

The studies discussed in this paper show that a simplified but adequately qualified formula set provides very satisfactory results. When applied to determining the input balance by the gravimetric method it presents the advantage of reducing the required number of neodymium analyses, and provides several independent values for checking the accuracy of the results.

The experience gathered from the first two water reactor fuel reprocessing campaigns at La Hague, and summarized in this paper, is extremely promising and confirms the potential interest of such methods. Other applications in the area of -non-destructive controls should also facilitate the enactment of nuclear material safeguards. Hull inspection by $^{144}Ce$ measurements is already practiced at La Hague, and fuel assembly identification, while not yet applied on an industrial scale, seems to present no unsurmountable technical difficulties.

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TABLE I

PRECISION OF "REACTOR" CALCULATIONS FOR A FUEL ASSEMBLY WITH AN ADJUSTED METHOD

| PARAMETERS \ PRECISION OF SPECIFIC BURNUP DATA | 5 % | 1 % |
|---|---|---|
| $^{235}U$ depletion | 5 % | 1 à 1,5 % |
| $^{236}U$ formation | 6 à 7 % | 2 % |
| Pu/U ratio | 3 à 5 % | 2 % |
| Pu isotopic abundance (240,241,242) | 5 à 10 % | 3 à 5 % |

TABLE II

MEASUREMENT - CALCULATION DEVIATION PERCENTAGE BEFORE ADJUSTMENT

(Fuel sample from the Ardennes Nuclear Plant)

| Mean specific burnup (MWD/T) | 12000 MWJ/T | 20000 MWJ/T | 25000 MWJ/T |
|---|---|---|---|
| $\Delta$ ( $U^{235}$ ) | + 1.5 | + 0.8 | + 2.5 |
| $U^{236}/U^{230}$ | - 2.0 | - 0.8 | - 0.5 |
| $Pu^{239}/ U^{238}$ | - 5.2 | - 4.2 | - 7.1 |
| $Pu^{241}/Pu^{239}$ | + 3.5 | + 2.3 | + 1.7 |
| $Pu^{242}/Pu^{239}$ | + 1.5 | + 1.0 | + 2.1 |

TABLE III

MEASUREMENT - CALCULATION DEVIATION PERCENTAGE AFTER ADJUSTMENT

| | Ardennes* | Stade** |
|---|---|---|
| $\Delta\ (U^{235})$ | + 0.7 | - 0.4 |
| $U^{236}/U^{238}$ | - 1.0 | + 0.1 |
| $Pu^{239}/\ U^{238}$ | - 0.6 | - 0.4 |
| $Pu^{240}/Pu^{239}$ | + 1.3 | + 0.8 |

* Measurement results for fuel rod samples

** Measurement results for samples from the dissolver at La Hague

TABLE IV

CORRELATION SENSITIVITIES TO SPECTRUM PERTURBATION DURING IRRADIATION AND TO UNCERTAINTY WITH REGARD TO FUEL HISTORY DATA

| Influence on relation / Perturbation | | | A | B | C | D |
|---|---|---|---|---|---|---|
| TCF | ⟷ | Δ (U5) | 3 % | 5 % | 2 % | 2 % |
| TCF | ⟷ | Δ (U6) | 1 % | 5 % | 0 | 1 % |
| TCF | ⟷ | Pu/U | 0 | 5 % | 6 % | 8 % |
| Pu/U | ⟷ | Pu9/Pu0 | 0,5 % | 10 % | 3 % | 5 % |
| Pu/U | ⟷ | $\frac{Pu2/Pu0}{Pu0/Pu9}$ | 0 | 1 % | 2 % | 4 % |
| Pu/U | ⟷ | $\frac{Pu - Pu9}{Pu}$ | 0,3 % | 0 | 7 % | 11 % |

REFERENCE : PWR Zr type 17 X 17

li = 3.2 %

I = 24 000 MWD/T

PERTURBATION

A - 1 % error on initial enrichment

B - 10 % error on declared specific burnup (without iteration)
In practice, the consequences of this uncertainty are negligible if the specific burnup is iterated

C - Partial poisoning of the fuel assembly during irradiation (Pyrex)

D - 5 % variation from mean moderation value

TABLE V

SPECIFIC BURNUP DETERMINATION

Comparison of Methods (Student's test) (8)

| METHODS COMPARED | $\lvert\bar{D}\rvert$ | | | s D | | | Student's test | | |
|---|---|---|---|---|---|---|---|---|---|
| | CNA | MUL | STA | CNA | MUL | STA | CNA | MUL | STA |
| R-Δ(U5) | - | 0.15 | 0.33 | - | 1.52 | 1.46 | 0 | - | 0.33 |
| R-Δ(U6) | - | 0.41 | 0.31 | - | 3.24 | 2.02 | 0 | - | - |
| R-Pu/U | - | - | 0.30 | - | - | 2.85 | 0 | 0 | - |
| Δ(U5)-Δ(U6) | 0.02 | 0.27 | 0.26 | 1.96 | 3.51 | 2.08 | - | - | - |
| Δ(U5)-Pu/U | - | - | 0.63 | - | - | 2.88 | 0 | 0 | - |
| Δ(U6)-Pu/U | - | - | 0.61 | - | - | 2.56 | 0 | 0 | 0.61 |
| Nd-Δ(U5) | 0.68 | 1.05 | 0.04 | 1.27 | 2.15 | 1.75 | 0.68 | - | - |
| Nd-Δ(U6) | 0.66 | 1.72 | 0.92 | 1.37 | 3.00 | 1.91 | 0.66 | - | - |
| Nd-Pu/U | - | - | 0.69 | - | - | 3.35 | 0 | 0 | - |
| Nd - R | - | 0.91 | 0.14 | - | 1.98 | 1.64 | 0 | - | - |
| Nd - Cs | - | 0.59 | - | - | 2.95 | - | 0 | - | - |

KEY :

CNA : Ardennes Nuclear Plant PWR (33 measurements on fuel rod samples)

MUL : Muhleberg BWR (17 measurements on solutions at La Hague)

STA : Stade PWR (81 measurements on solutions at La Hague, except for Nd : 15 values only)

$\bar{D}$ : Mean deviation between methods

sD : Standard deviation on $\bar{D}$

Student's Test : 0 = no test

\- = no systematic deviation

or most probable systematic deviation value

METHODS :

R Specific burnup announced by reactor operator

(U5) Specific burnup calculated from $^{235}U$ depletion

(U6) Specific burnup calculated from $^{236}U$ formation

Pu/U Specific burnup calculated from plutonium formation

Nd Specific burnup calculated from neodymium measurement.

TABLE VI

SPECIFIC BURNUP DETERMINATION

Random Error Estimate (9)

| METHOD | STANDARD DEVIATION | | | |
|---|---|---|---|---|
| | CNA 30 Measurm. | MUL 14 measurm. | STADE 15 measurm. | STADE 81 measurm. |
| | - | 0.4 | 1.1 | 1.3 |
| | 0.0 | 1.4 | 1.3 | - |
| Δ (U5) | 1.3 | 1.3 | 1.0 | 1.3 |
| Δ (U6) | 1.5 | 2.9 | 1.1 | 1.5 |
| Pu / U | - | - | 3.1 | 2.5 |
| Césium | - | 2.3 | - | - |

KEY : Refer to Table V

TABLE VII

Pu/U RATIO DETERMINATION FOR STADE PWR (81 measurements)

| COMPARISON OF METHODS | | | RANDOM ERROR ESTIMATE | |
|---|---|---|---|---|
| METHODS COMPARED | $\bar{D}$ (%) | $\Delta D$ (%) | METHOD | $\sigma$ (%) |
| Mesure - Reactor | - 1.85 | 2.46 | Mesure | 0.4 |
| Mesure - TCF | 0.37 | 1.50 | Reactor | 2.9 |
| Mesure - Pu9 / Pu0 | 1.17 | 1.79 | TCF | 1.1 |
| Mesure - $\frac{Pu9 / Pu0}{Pu0 / Pu9}$ | 5.16 | 1.98 | Pu9/Pu0 | 1.7 |
| Reactor - TCF | 2.25 | 3.10 | $\frac{Pu9/Pu0}{Pu0/Pu9}$ | 1.0 |
| Reactor - Pu9 / Pu0 | 3.02 | 3.42 | | |
| Reactor - $\frac{Pu2 / Pu0}{Pu0 / Pu9}$ | 6.86 | 2.12 | | |
| T C F - Pu9 / Pu0 | 0.80 | 0.85 | | |
| T C F - $\frac{Pu2 / Pu0}{Pu0 / Pu9}$ | 4.64 | 1.80 | | |
| Pu9/Pu0 - $\frac{Pu2 / Pu0}{Pu0 / Pu9}$ | 3.84 | 2.16 | | |

KEY :

$\bar{D}$ Mean deviation between methods

$\Delta D$ Standard deviation on $\bar{D}$

$\sigma$ Estimated standard deviation per method

Measurement Direct Pu/U ratio determination by mass spectrometry

Reactor Estimated Pu/U ratio supplied by reactor operator

TCF Pu/U ratio calculated from measured specific burnup

Pu9/Pu0 Pu/U ratio calculated from measured Pu9/Pu0 isotopic ratio

$\frac{Pu2/Pu0}{Pu0/Pu9}$ Pu/U ratio calculated from measured $\frac{Pu2/Pu0}{Pu0/Pu9}$ isotopic ratio

TABLE VIII

URANIUM MASS DETERMINATION FOR STADE PWR

a - Comparison of Methods

| SPECIFIC BURNUP DETERMINATION METHOD | DEVIATION PERCENTAGE FOR URANIUM MASS | | | | | |
|---|---|---|---|---|---|---|
| | $\lvert \bar{D} \rvert$ | | s D | | TEST | |
| | 15 Meas. | 81 Meas. | 15 Meas. | 81 Meas. | 15 Meas. | 81 Meas. |
| Nd - Reactor | 0.02 | - | 0.11 | - | - | 0 |
| Nd - Δ (U5) | 0.01 | - | 0.12 | - | - | 0 |
| Nd - Pu/U | 0.02 | - | 0.15 | - | - | 0 |
| Reactor - Δ(U5) | 0.03 | 0.03 | 0.05 | 0.07 | - | 0.03 |
| Reactor - Pu/U | 0.00 | 0.02 | 0.11 | 0.09 | - | - |
| Δ (U5) - Pu/U | 0.03 | 0.05 | 0.12 | 0.11 | - | 0.05 |

b - Random Error Estimate

| METHOD | STANDARD DEVIATION | |
|---|---|---|
| | 15 measurements | 81 measurements |
| Neodyme | 0.03 | - |
| Reactor | 0.03 | 0.04 |
| Δ (U5) | 0.04 | 0.06 |
| Pu/U | 0.10 | 0.09 |

KEY : Refer to Table V

TABLE IX

EXAMPLES OF $\frac{^{134}Cs}{^{137}Cs}$ AND $\frac{^{144}Ce}{^{137}Cs}$ RATIO SENSITIVITY TO

FUEL IRRADIATION HISTORY

| CHRONOLOGY | OPERATING POWER LEVEL (%) OF NOMINAL | | | | |
|---|---|---|---|---|---|
| | REFERENCE | 1 | 2 | 3 | 4 |
| 0 | 75 % | 75 % | 75 % | 75 % | 75 % |
| 1 year | 75 % | 75 % | 75 % | 50 %<br>100 % | 75 % |
| 2 years | 75 % | 100 %<br>50 % | 50 %<br>100 %<br>50 % | 75 % | 0<br>75 % |
| 3 years | Defueling | Defueling | Defueling | Defueling | Defueling |

| ISOTOPIC RATIO DEVIATION PERCENTAGE WITH RESPECT TO REFERENCE HISTORY AS OF DEFUELING DATE | | | | | |
|---|---|---|---|---|---|
| $^{134}Cs / ^{137}Ce$ . | 0 | - 1.8 | - 0.1 | 0 | - 3.6 |
| $^{144}Ce / ^{137}Cs$ | 0 | - 4.6 | - 0.5 | + 0;2 | - 10.4 |

TABLE X

FUEL IRRADIATION HISTORY SENSITIVITY OF THE CORRELATION BETWEEN FUEL MASS AND VARIOUS ACTIVITY LEVELS

| PERTURBATION / EFFECT ON CORRELATION BETWEEN FUEL MASS AND : | A | B | C | D |
|---|---|---|---|---|
| Total gamma activity | 5 % | 5 % | 5 % | 0,5 % |
| Total neutron activity | 50 % | 3 % | 1 % | 16 % |
| $^{137}Cs$ gamma activity | 10 % | 0,2 % | 0,2 % | 0 |
| $^{136}Cs/^{137}Cs$ gamma activity | 8 % | 3,5 % | 2,5 % | 3,5 % |
| $^{144}Ce$ gamma activity | 1,5 % | 7 % | 7 % | 1 % |

REFERENCE

PWR - Zr 17x17

Li = 3.2 %

I = 24000 MWD/T

Cooling time = 1 year

PERTURBATIONS

A - 10 % error on specific burnup estimate

B - 10 % error on cooling time

C - 10 % error on irradiation time at mean steady-state power

D - 10 % error on initial enrichment.

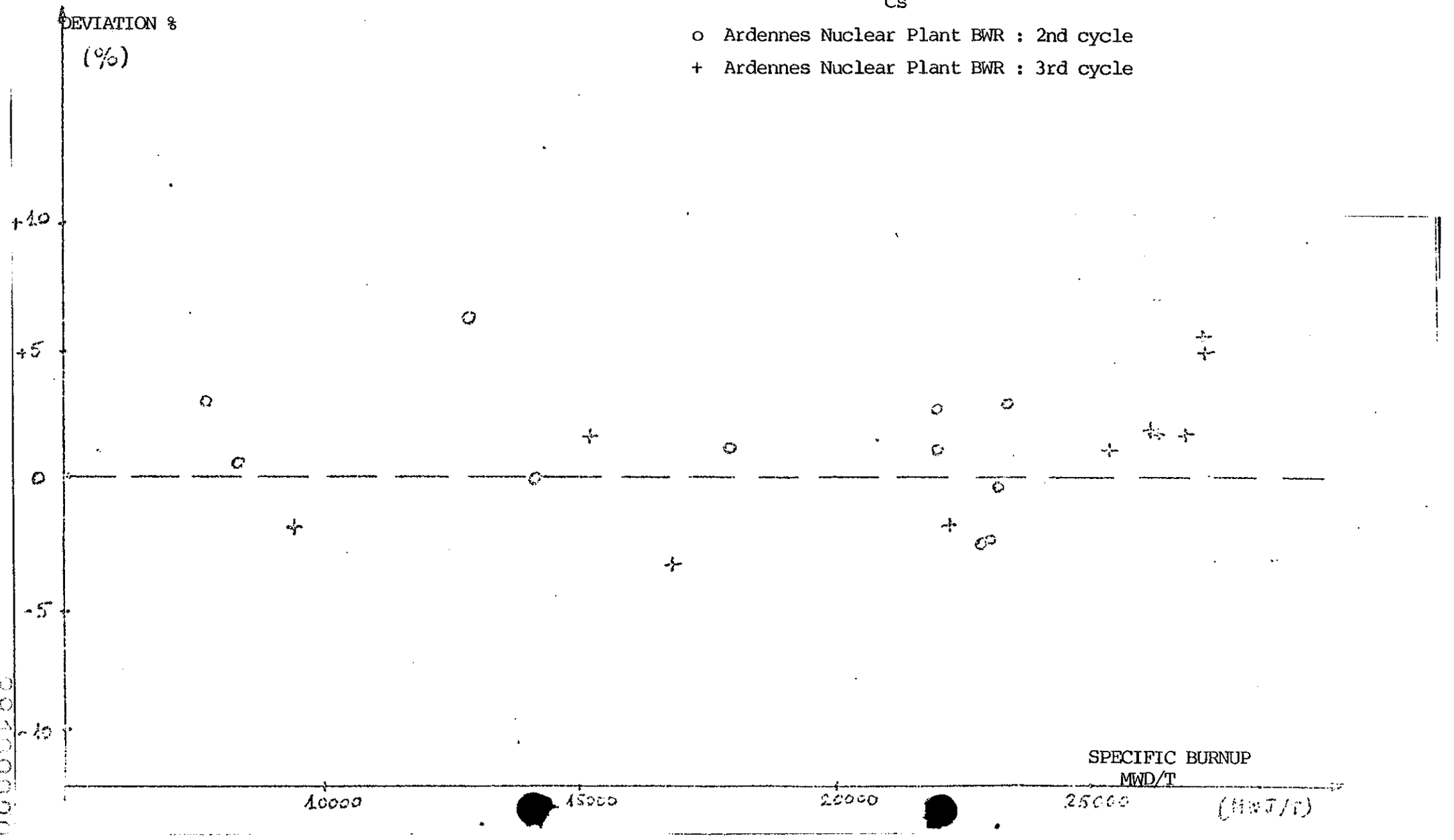
FIGURE 1

COMPARISON OF SPECIFIC BURNUP VALUES

CALCULATED FROM $\frac{^{134}Cs}{^{137}Cs}$ ACTIVITY RATIO AND Nd CONTENT

o Ardennes Nuclear Plant BWR : 2nd cycle

+ Ardennes Nuclear Plant BWR : 3rd cycle



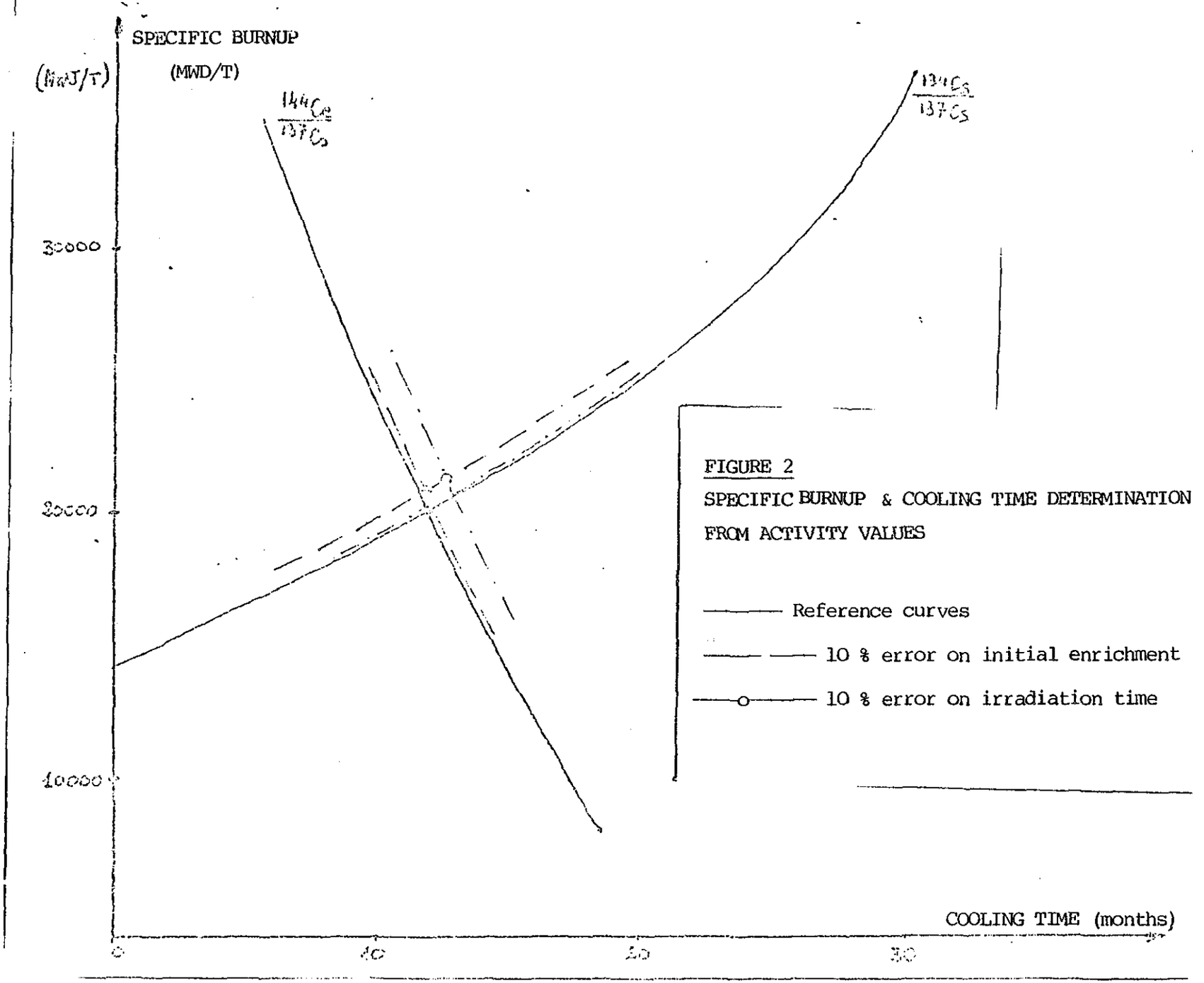



FIGURE 2

SPECIFIC BURNUP & COOLING TIME DETERMINATION FROM ACTIVITY VALUES

——— Reference curves

——— ——— 10 % error on initial enrichment

———o——— 10 % error on irradiation time

FIGURE 3

$^{144}Ce$ ACTIVITY VERSUS SPECIFIC BURNUP AND COOLING TIME : NOMOGRAMS FOR HULL CONTROL