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SECONDARY ACTINIDE FORMATION & DECAY IN LIGHT WATER & FAST BREEDER REACTORS

REPERCUSSIONS ON THE FUEL CYCLE

M. DARROUZET-A. GIACOMETTI-M. ROBIN

Light Water Reactor Department
Nuclear Research Service

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COMMISSARIAT A L'ENERGIE ATOMIQUE
Centre d'Etudes Nucléaires de Cadarache
13115 SAINT-PAUL-LEZ-DURANCE
FRANCE

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INTRODUCTION

The rapid expansion of the French nuclear power program and the effort to optimize the nuclear fuel cycle have highlighted a number of problems previously considered marginal.

With increasingly higher burnup values and with uranium and plutonium recycling to save reserve supplies, it is no longer possible to disregard the "secondary" actinides i.e. the actinides other than the major uranium and plutonium isotopes. These elements modify the reactor neutron balance, result in substantially higher radioactive doses during transport, in reprocessing facilities and in fuel fabrication plants, and raise the problem of massive accumulation in time.

It is therefore increasingly necessary to develop reliable calculation methods capable of determining, within acceptable accuracy limits, the effects of these actinides on all the steps of the fuel cycle.

The research work undertaken in this area by the CEA more specifically concerns the French nuclear power options, i.e. PWR and fast breeder reactor development, spent fuel reprocessing, uranium enrichment, uranium oxide or uranium-plutonium mixed oxide fuel fabrication.

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1 - SECONDARY ACTINIDE FORMATION IN REACTORS (1)

1.1. Actinide Formation

Table I shows the nuclides with half-lives exceeding one day which are found in reactors fueled initially with uranium or its daughter product plutonium. The table shows the major processes by which secondary actinides are formed from nuclides which are essential to reactor operation. These nuclides are formed by neutron capture (n, γ reaction), radioactive decay (α , β , electron capture) and by (n, 2n) reaction. All other formation processes may be disregarded.

1.2 Calculation Methods

Secondary actinide concentrations cannot be calculated independently of the primary actinide values (uranium and plutonium). Moreover, the flux history must be considered in detail since short half-life nuclides are involved in many transmutation series.

Secondary actinide calculations are performed with reactor computer codes which use multigroup cross sections with allowance for neutron spectra and which are capable of calculating actinide concentrations : the NEPTUNE system (2) using the APOLLO cell code for thermal reactors, and the CARNAVAL set (3) using the HETAIRE cell code for fast neutron reactors. These codes have been qualified repeatedly for the primary nuclei.

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Experience has shown, however, that these codes are not perfectly suited to secondary actinide studies. They do not always provide adequate flexibility to handle certain formation processes (n, 2n reaction, α decay, etc.) and are expensive to use when all the secondary actinides are to be taken into account together with the detailed history of neutron flux variations.

The neutron spectrum and the primary isotope variation are not altered by the presence of most secondary actinides, so that it is feasible to calculate the concentration of these secondary actinides using computer codes coupled with the reactor codes.

The EVOGENE code was developped for this purpose. It is capable of calculating concentration variations for forty heavy isotopes and fifty fission products, taking all formation processes into account. It also determines the α activity, the residual power (following one month of cooling) and spontaneous neutron emission (spontaneous fissions and (α, n) reactions). The cross sections condensed to a single group over the flux values obtained from the reactor codes are introduced in parameter form as a function of the burnup (4).

This procedure provides accurate data on the primary actinides which constitute the starting point for the secondary actinide series. Moreover, the secondary actinide cross sections are thoroughly weighted in the relevant fuel spectrum. The flux history may be very closely taken into account.

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1.3 - Actinides Amounts Formed

Table II shows the secondary actinide masses present on defueling of three typical fuels :

- UO_2 fuel irradiated to 32000 MWd/MT in a PWR
- UO_2-PuO_2 fuel irradiated to 85000 MWd/MT of oxide in a fast neutron reactor, fabricated using :
  . plutonium from a PWR
  . plutonium recycled several times in the fast reactor.

The masses are expressed in grams per metric ton of initial oxide.

For several actinides, the concentrations vary significantly during cooling.
Thus, for example, after two years of cooling the amounts shown in Table II must be multiplied by the factors shown in Table III.

1.4 - Relative Importance of Actinide Formation Processes

Table I shows the major processes by which the secondary actinides are formed. Some actinides are formed in more than one manner ; minor processes have been intentionally omitted from this chart.

A number of precisions may be made concerning the importance of the various processes by which ^{237}Np, ^{238}Pu, ^{243}Am and ^{244}Cm are formed. Table IV shows the percentage breakdowns obtained at the moment of defueling. These calculations assume that the uranium initially contained no ^{236}U, and that 0.4 % depleted uranium was used in the fast reactor fuel.

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In light water reactors, ^{237}Np and ^{238}Pu are primarily formed from ^{235}U, while in fast breeder reactors the ^{237}Np is formed from ^{238}U, and the ^{238}Pu from ^{242}Cm decay.

Only minute amounts of ^{243}Am and ^{244}Cm are formed by (n, γ) reaction from ^{242}Am or ^{243}Cm.

2 - SECONDARY ACTINIDES IN THE FUEL CYCLE (5) (6)

2.1 Irradiation

The actinide reactivity value in the reactor depends on the amounts present in the core, as well as on the mean $\bar{\nu}$ values and cross sections.

The effects are slight ($<$ 1000 pcm), but this result is frequently due to shimming.

The pre-adjustment uncertainty range on the cross sections is estimated at $\pm$ 100 pcm is PWRs, and may reach $\pm$ 500 pcm if plutonium containing large amounts of heavy isotopes is used in FBRs.

The predominant effects are attributable to ^{237}Np in PWRs and to ^{238}Pu and Americium isotopes in fast reactors.

The short half-life actinides (e.g. ^{239}Np) cannot be disregarded — especially in fast neutron reactors — and must be taken into account during sudden power swings and in residual power calculations.

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2.2 Transport and Reprocessing

Here the problem involves alpha activity and neutron emission. Table V shows these activity values (per metric ton of initial oxide) both 3 months and 1 year after defueling : most of this activity comes from ^{242}Cm and ^{244}Cm.

The uncertainty bounds on these values due to inadequate data both on certain cross sections and on (α , n) yields, and may reach 50 to 60 % before adjustment.

Table V also indicates the residual power values. It may be seen that after one year the actinides account for 5 to 15 % of the total power released from the fuel.

2.3 - Fuel Fabrication with Plutonium or Recycled Uranium

The problems arising during the fuel fabrication stage, other than those involving criticality, result from the alpha and neutron activities and from the gamma doses.

For the latter, in particular, allowance must be made for γ emission not only from the actinide initially present but also from all its daughter nuclides.

No special problems arise for UO_2 fabrication, except where recycled uranium is used. The difficulty in this case stems from the 2.6MeV gamma emission by ^{208}Tl derived from ^{232}U.

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This activity thus depends both on the degree of uranium/plutonium separation achieved during reprocessing (^{232}U is formed by decay of ^{236}Pu, with a 2.8-year half-life) and on the uranium storage time prior to fabrication : ^{232}U has a 72-year half-life, while all the intermediate isotopes leading to ^{208}Tl formation have very short half-lives (less than 4 days) except for ^{228}Th, with a 1.9-year half-life.

The result in Table VI show that the γ doses on contact with a significant mass may be multiplied by a factor of 20 or more between the "natural" UO_2 and the recycled UO_2.

In the case of UO_2-PuO_2 fast reactor fuel, the problem is essentially due to plutonium (especially ^{238}Pu and ^{240}Pu) for the alpha and neutron activity, and to ^{241}Am for the gamma activity. The actual activity values vary according to the origin of the plutonium. Table VII shows the approximate magnitude of these values for plutonium from various types of reactors, 12 months after separation of the americium content. Here again, the ^{236}Pu and ^{232}U cannot by fully disregarded.

3 - PLUTONIUM RECYCLING IN LIGHT WATER REACTORS AND ACTINIDE RECYCLING IN FAST BREEDER REACTORS

3.1 - Plutonium Recycling in Light Water Reactors

The French nuclear program does not involve plutonium recycling in light water reactors, since large amounts of plutonium will be required by the development of fast breeder reactors. Some basic research has nevertheless been done in this area.

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Table VIII shows the amounts of secondary actinides formed ; these quantities are much greater (3-4 times higher) than in the applications discussed above.

Americium is much more significant in the neutron balance (by a factor of 10 compared to UO_2 fuel) ; the same is true of the activity values on defueling.

3.2 - Actinides Recycling in Fast Breeder Reactors

One means of eliminating the long half-life alpha-emitting actinides is to burn them in fast breeder reactors (5) (7).

Studies on recycling plutonium together with Am and Np isotopes have shown that the secondary actinide quantities reach saturation.

Table IX shows the amounts of these actinides present in the fuel on unloading after fifteen years of reactor operation. These amounts are not higher than with LWR plutonium from which the ^{237}Np and ^{241}Am are separated at the outset.

The alpha activity and neutron emission levels of the fuel on fabrication are quite acceptable. The γ activity is substantially higher than when the Am and Np isotopes are not recycled.

Curium recycling seems at present much less favorable. The amount of curium in the core does not saturate, and fuel activity levels are much higher on fabrication (refer to Tables IX and X).

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4 - EXPERIMENTAL PROGRAM

A major CEA research program is under way to qualify the calculations for secondary actinide formation and the resulting effects. This work essentially involves :

- evaluating cross sections
- compiling non-neutron nuclear data on heavy nuclei (8)

Together with these studies, an experimental program has been undertaken for several years with the intention of improving assessments of effective cross sections for the secondary actinides in PWRs and FBRs, of testing the isotope concentration calculations for spent fuel and of checking the γ dosage and activity estimates for various types of fuel at the fabrication stage.

This program comprises three basic points :

- analysis of fuel or separated isotope samples
- physical measurements in critical experiments
- control of neutron emission and γ radiation.

4.1 - Spent Fuel Analysis

An overall control of the calculation is possible on the basis of spent power reactor fuel analyses, but it is difficult to determine the adjustments required for the cross section libraries used.

Analyses of separated isotope samples irradiated in known spectra provide precise determinations of the integral capture cross sections, but require special irradiations.

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Experimental techniques had to be developed for the secondary actinides. The americium and curium isotopic contents and the Am/Pu ratio (double isotopic dilution) are measured by mass spectrometry. The ^{232}U, ^{242}Cm and ^{244}Cm contents are measured by alpha spectrometry (after uranium ultrapurification for the ^{232}U) and a ^{233}U - ^{239}Np tracer is used for ^{237}Np. The accuracy of these measurements is highly dependent on the nuclide quantities present, i.e. on the burnup.

These studies have concentrated on the two major reactor systems in use in France.

a) Light Water Reactors

This program was based primarily on analyses of fuel unloaded after the first three operating cycles of the Ardennes nuclear plant, and especially pins situated in "neutronically clean" conditions. The burnup values ranged from 8000 MWd/MT to 35000 MWd/MT. The fuel in this reactor is contained in steel cladding.

Table XI shows the mean deviation and dispersion values on the computed and experimental results for the primary isotopes in 23 samples, while Table XII presents the relevant values for the secondary actinides.

The experimental/calculation deviations were occasionally very significant with the first cross section library used for the secondary actinides. Adjustments were accordingly introduced to substantially reduce these discrepancies, and the resulting library should be free of any major errors on secondary actinide calculations.

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Further checks are scheduled for PWR fuel unloaded from the Tihange plant (15 X 15, zircaloy cladding) and the Fessenheim plant (17 X 17, zircaloy cladding).

Moreover, individual isotopes (^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm) are to be irradiated separately in the center of a 5 X 5 rod array in the Melusine reactor under neutron conditions similar to those of a PWR power plant. The actinides will be introduced into the UO_2 fuel pellets at about 1 % abundance to simplify interpretation.

This experiment will be undertaken in the scope of the Euratom research program on plutonium recycling, and will substantially diminish the cross section uncertainty bounds for these nuclides.

b) Fast Breeder Reactors

An experimental program is now in progress on fuel discharged at different burnup levels from Phénix cores 1 and 2 (Pu and U assemblies). Analysis results are also available for separate isotope samples irradiated in Phénix (the PROFIL experiment). Table XIII shows, referenced to ^{238}U, the capture ratio deviations between measurements and the values calculated using the CARNAVAL III library. The primary objective of this initial irradiation was to determine the capture ratios for the major uranium and plutonium isotopes.

A second experiment (PROFIL II) is planned more specially for the secondary actinides, and should significantly reduce present uncertainty as to the capture ratios for these isotopes.

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4.2 - Physical Measurements in Critical Experiments

a) Fission Chamber Measurements

Irradiation measurements are used to confirm the capture ratio values. A highly accurate method of obtaining fission rate data is to use miniature fission chambers for the measurements. This technique, commonly used for the major uranium and plutonium isotopes, has been adapted to the strong alpha-emitting secondary actinides (9).

In the scope of the fission chamber measurement program on fast breeder reactors, the fission cross sections of ^{238}Pu and ^{241}Am were determined in various lattices during the ERMINE coupled thermal/fast neutron experiment. These results are used to correct the cross sections to within $\pm$ 5 % in the spectrum range relevant to fast breeder reactors.

Figure 1, for example, plots the deviations between the experimental results and the values calculated using CARNAVAL III (before adjustment) for the σ_{f241Am} σ_{f235U} ratio versus the spectral hardness parameter $r = \frac{\nu\Sigma f}{\xi\Sigma s}$

This type of measurement continues to be made, and will be used in particular for ^{243}Am and ^{244}Cm.

b) Activation Measurements

Short-term irradiation capture ratio measurements are planned for isotopes which by capture produce a short half-life isotope emitting a readily detectable gamma radiation , in particular ^{236}U, ^{237}Np and ^{243}Am.

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4.3 - Emission Control

Calculated values for fuel neutron emission are checked during measurement campaigns on standard control samples using the PRIMEVERE system.

Table XIV shows the $\frac{\text{experiment-calculation}}{\text{calculation}}$ deviations recorded for plutonium samples containing various concentrations of heavy isotopes and of ^{238}Pu.

Other measurements are conducted in conjunction with the fabrication facilities as a general check of neutron and gamma dose calculations for various types of fuels.

CONCLUSION

The problem of the secondary actinides and the resulting radiation emissions (α , γ ,neutrons) throughout the fuel cycle is covered by a substantial CEA research effort. Computer methods are now available to handle both existing problems and planned studies.

The experimental program to date has significantly narrowed the uncertainty bounds on all the major parameters relevant to the secondary actinides.

The studies now under way will provide further valuable data both for reactor project design and for fuel reprocessing and fabrication plant operation.

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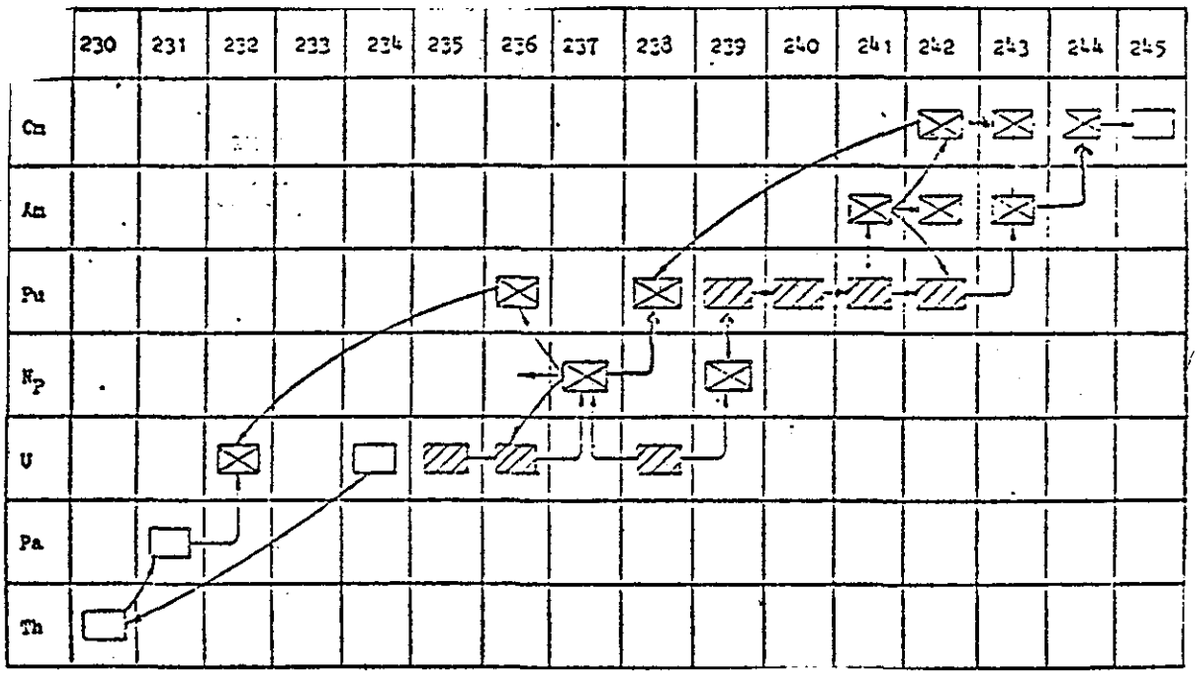
TABLE I

"FORMATION OF THE SECONDARY ACTINIDES"



▨ Primary actinides

⊠ Secondary actinides studied

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TABLE II

"MASS (grams per initial oxide metric ton) OF SECONDARY ACTINIDES IN SPENT FUEL"

REACTOR TYPE	PWR*	FBR** UO_2-PuO_2	
FUEL TYPE	UO_2	"LWR Pu"	"Equilibrium Pu"
Isotope			
232 U	0.8×10^{-3}	1.6×10^{-2}	5.7×10^{-3}
237 Np	450	290	275
236 Pu	2.2×10^{-3}	17×10^{-3}	13×10^{-3}
238 Pu	155	1510	545
241 Am	35	1100	560
242m Am	0.6	126	50
243 Am	100	850	580
242 Cm	14	145	60
243 Cm	0.5	13	4
244 Cm	26	240	160
245 Cm	0.15	15	10

* 32 000 MWd/MTo

** 85 000 MWd/MTo

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TABLE III

"MASS RATIO OF ACTINIDES PRESENT IN SPENT FUEL AFTER 2 YEARS OF COOLING TO ACTINIDES PRESENT AT THE TIME OF DEFUELING"

REACTOR TYPE	PWR	FBR UO_2-PuO_2	
FUEL TYPE	UO_2	"LWR Pu"	"Equilibrium Pu"
Isotope			
232 U	2.0	1.4	1.9
237 Np	1	1	1
236 Pu	0.6	0.6	0.6
238 Pu	1	0.98	0.98
241 Am	4.2	1.5	1.9
242m Am	1	1	1
243 Am	1	1	1
242 Cm	4.5×10^{-2}	4.5×10^{-2}	4.5×10^{-2}
243 Cm	1	1	1
244 Cm	0.93	0.93	0.93
245 Cm	1	1	1

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TABLE IV

"ACTINIDE FORMATION PERCENTAGE BREAKDOWN"

ISOTOPE	FORMATION PROCESS	PWR (UO_2)	FBR (LWR Pu)	FBR (Equilibrium Pu)
		32000MWd/MTo	85000 MWd/MTo	
^{237}Np	(n,γ) from ^{236}U	80	14	10
	n,2n from ^{238}U	20	86	90
^{238}Pu	(n,γ) from ^{237}Np	91	15	29
	(α) from ^{242}Cm	9	73	56
	(n,2n) from ^{239}Pu	< 0.1	12	15
^{243}Am	(n,γ) from ^{242}Pu	99.4	95.4	97.4
	(n,γ) from ^{242}Am	0.6	4.6	2.8
^{244}Cm	(n,γ) from ^{243}Am	99.6	99.6	99.8
	(n,γ) from ^{243}Cm	0.4	0.4	0.2

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TABLE V

"ALPHA & NEUTRON ACTIVITY LEVELS AND RESIDUAL POWER IN SPENT FUEL"

REACTOR TYPE	FUEL	COOLING TIME (months)	α ACTIVITY (kCi/MTo)	NEUTRON EMISSION (10^9 n/sec/MTo)	POWER FROM ACTINIDES (kW/MTo)	POWER FROM FISSION PRODUCT (kW/MTo)
PWR	UO_2	3	36	0.55	1.30	27
		12	16	0.38	0.55	11
FBR	LWR Pu	3	340	4.7	12	80
		12	150	3.2	5.3	30
	Equilibrium Pu	3	170	2.7	6.0	80
		12	80	2.0	2.8	30

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TABLE VI

"3.2 % ENRICHED URANIUM FUEL FABRICATION : ACTIVITY PER gUO_2"

ORIGIN	ACTIVITY	NEUTRON EMISSION	γ DOSE*
Natural U	2 µCi/g	10^{-2}n/g	2 mrem/h
U recycled after 1-year cooling	6 µCi/g	10^{-2}n/g	40 mrem/h

* Dose on contact with a large quantity of UO_2

TABLE VII

"PLUTONIUM FUEL FABRICATION : ACTIVITY PER g PuO_2"

REACTOR TYPE	ACTIVITY mCi/gPuO_2	NEUTRON EMISSION (spontaneous fission + α,n) n/sec/g PuO_2	γ DOSE* rem/h
Graphite-gas : UO_2	95	275	1.70
PWR : UO_2	340	630	4.30
FBR : "équilibrium"	140	365	1.35

* Dose on contact with a large quantity of PuO_2

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TABLE VIII

"MASS (grams per initial oxide metric ton) OF SECONDARY ACTINIDES IN UO_2-PuO_2 FUEL AT THE MOMENT OF UNLOADING FROM A PWR AFTER IRRADIATION TO 32000 MWd/MT"

^{232}U	6.8×10^{-3}	242m Am	14
^{237}Np	200	^{243}Am	1235
^{236}Pu	7.4×10^{-3}	^{242}Cm	130
^{238}Pu	965	^{243}Cm	8
^{241}Am	565	^{244}Cm	440

TABLE IX

"MASS OF SECONDARY ACTINIDES AFTER RECYCLING IN SPENT FUEL AT THE MOMENT OF UNLOADING FROM AN FBR"

	LWP Pu	RECYCLING			
		Equilibrium Pu	Pu + Np	Pu + Am	Pu + Np + Am + Cm
Np	290	275	565	275	565
Am	2076	1190	1190	2525	2525
Cm	413	235	235	819	1460

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TABLE X

"PLUTONIUM FUEL FABRICATION
ACTIVITY PER $gPuO_2$ ASSUMING ACTINIDES RECYCLED WITH Pu"

	α ACTIVITY mCi/g	NEUTRON EMISSION n/sec/g	γ DOSE rem/h
Pu + Np	167	390	1.4
Pu + Am	212	475	34
Pu + Np + Am	750	62 000	52

TABLE XI

"LIGHT WATER REACTORS : PRIMARY ACTINIDE DATA
$\frac{\text{EXPERIMENT - CALCULATION}}{\text{CALCULATION}}$ PERCENTAGE DEVIATIONS"

ISOTOPE RATIO	$\frac{E - C}{C}$	DISPERSION
235 U/238U	+ 0.5	± 3
239Pu/238U	+ 0.5	± 5
240Pu/239Pu	+ 4	± 4
241Pu/239Pu	+ 0.5	± 3

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TABLE XII

"LIGHT WATER REACTORS : PRIMARY ACTINIDE DATA EXPERIMENT - CALCULATION / CALCULATION PERCENTAGE DEVIATIONS"

ISOTOPE RATIO	NUMBER OF MEASUREMENTS	LIBRARY PRE-ADJUSTMENT	LIBRARY POST-ADJUSTMENT	DISPERSION
236U/238U	23	+ 7	+ 0.4	± 5
237Np/238U	4	-10	- 4	± 6
232U/238U	3	+ 6	+ 7	±15
238Pu/239Pu	18	+16	- 3	±16
242Pu/239Pu	23	-10	- 5	± 6
242Cm/239Pu	16	+12	+ 4	±16
244Cm/239Pu	16	+53	- 3	±28

TABLE XIII

"PHENIX FBR : CAPTURE RATIO DATA EXPERIMENT - CALCULATION / CALCULATION PERCENTAGE DEVIATIONS"

237 Np	+ 32 % ± 15 %	241 Am	- 8 % ± 12 %
239 Np	- 5 % ± 19 %	243 Am	- 10 % ± 10 %
238 Pu	- 22 % ± 3 %	244 Cm	- 19 % ± 15 %
242 Pu	- 36 % ± 5 %	(n, 2n) from 239 Pu	- 55 % ± 4 %

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TABLE XIV

"NEUTRON EMISSION (spontaneous fissions + α,n)
REFERENCED TO 1g of (Pu + Am)

	MEASURED VALUE (Arbitrary units)	$\frac{E - C}{C}$ (%)
Pu metal	2. 52	+ 2.9
	2.60	+ 6.6
	12.52	+ 5.2
	12.63	+ 6.0
	36.31	+ 8.0
Pu oxide	7.64	+ 1.0
	21.91	- 1.7
	33.72	+ 1.2
	62.28	- 1.2
	85.48	- 1.9
PuO_2 - UO_2 high Pu content > 15%	18.32	0.3
	18.20	- 0.3
	23.07	- 0.5
	101.30	- 1.5
	101.25	- 1.6
PuO_2 - UO_2 low Pu content < 6 %	11.82	- 1.6
	36.83	- 0.0
	37.82	- 1.9

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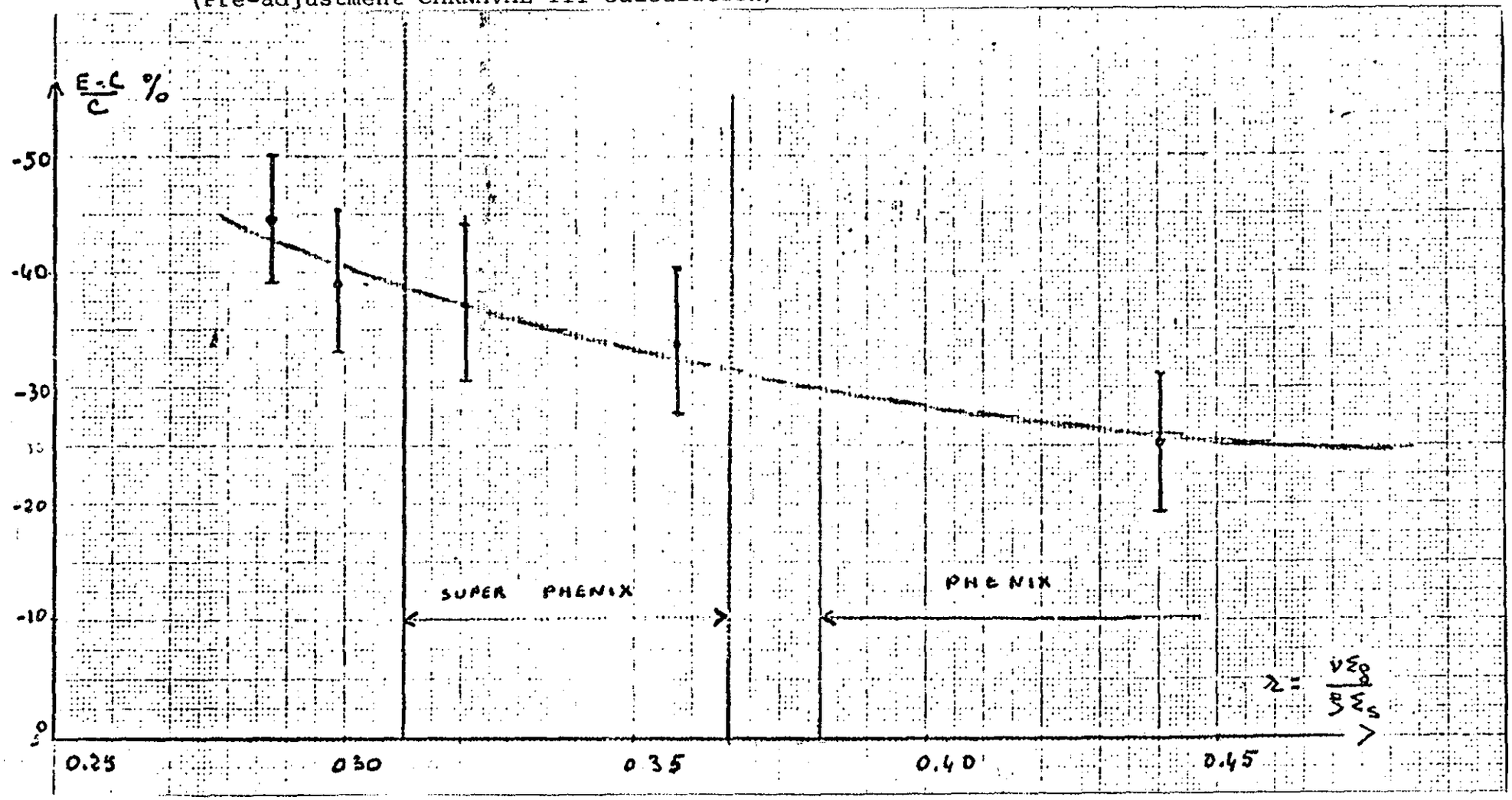
FIGURE 1 : FISSION INDEX: $^{241}Am/^{235}Pu$ $\frac{\text{EXPERIMENTAL - CALCULATION}}{\text{CALCULATION}}$ PERCENTAGE DEVIATION

(Pre-adjustment CARNAVAL III calculation)"



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