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TIME-DEPENDENT FISSION-PRODUCT THERMAL AND RESONANCE ABSORPTION CROSS SECTIONS

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BETTIS ATOMIC POWER LABORATORY, PITTSBURGH, PA.,
OPERATED FOR THE U. S. ATOMIC ENERGY COMMISSION
BY WESTINGHOUSE ELECTRIC CORPORATION

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RESONANCE ABSORPTION CROSS SECTIONS

T. R. England

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With the advent of increasing reactor lifetimes, the need for an accurate description of all fission product cross sections developed in an operating reactor is apparent. An exact analytical method is outlined by which the detailed time-dependent concentration of each fission product and other generated nuclides can be calculated. Radioactive decay and neutron absorption in the time-dependent reactor flux are explicitly included. In addition to U-235 several fissile nuclides may simultaneously provide a source to the fragment concentrations. The method is demonstrated by calculating both the individual and total thermal and resonance cross sections per fission of 163 fission products from U-235, natural uranium, enriched uranium, and Pu-239 in a variety of irradiation flux histories and spectra. All known fission products of significance, i.e., those formed by direct fission yields and their progeny formed by neutron absorption and radioactive decay, are included in the analysis. This is in contrast to treatments heretofore which have largely ignored coupling systematics. A comparison of the aggregate absorption cross sections, determined by both calculation and experiment for a highly enriched sample, which has been irradiated in the MTR to 81 percent U-235 depletion, shows agreement within 2 percent thermally and 15 percent epithermally.

TIME-DEPENDENT FISSION-PRODUCT THERMAL AND RESONANCE ABSORPTION CROSS SECTIONS

T. R. England

INTRODUCTION

This report presents the results of a study of the time-dependent, macroscopic absorbing properties of the nuclides created by the fission process. With the advent of the design of second generation nuclear reactors, many with substantially increased lifetime objectives, the need for an accurate description of the time-dependent concentrations of the fission products becomes apparent. Several authors (References 1 through 5) have recently studied the subject and presented comprehensive summaries of nuclide data along with various approximate treatments of fission product absorption or "poisoning" effects. Primarily, this study was motivated by a desire to obtain an adequate poison calculation for a seed-blanket reactor (PWR), which requires both an analysis of the progeny of U-235 fission (seed) and those of U-238, Pu-239, and Pu-241 (blanket). It was found that a treatment adequate for study of the natural uranium fuel in the blanket did not exist. Also and most important, simplifying assumptions were made by the various authors in all treatments of U-235 products, and the possible consequences of these for reactor lifetime and reactivity control studies were unknown. The most outstanding of these simplifications is the neglect of radioactive decay and its effect on the concentration of both the stable and radioactive, neutron-absorbing nuclei. Some authors neglected the nuclides formed by neutron capture in the primary fission products, and no treatment, heretofore, accounted for all coupling systematics and the formation of very high order products. Examination of the existing treatments indicated that substantial adjustments would be necessary to adapt their results to our depletion programs, and the validity of the approximate calculational schemes was unknown.

Before outlining the content of this study, an examination of the physical situation will be helpful.

The nuclides formed directly by the fission process appear in about 79 isobaric chains. Most of these nuclides are unstable, primarily with respect to β -decay. Thus, in the absence of a neutron flux the behavior of the fission products would be governed solely by the transmutation of the β -emitters which eventually decay to stable nuclides. Therefore, the products of fission could be grouped into a set

of independent isobaric chains, each of which could be rather easily described since each radioactive isobar would possess a unique decay path from one unstable product to another, and the chain would finally terminate in a stable nucleus. Examination of the physical data of the radioactive direct products indicates that the 79 chains would each contain three β -coupled nuclides on the average.

In a reactor environment the situation becomes vastly more complex. The stable and unstable nuclides absorb neutrons, and this cross-links or couples the isobaric chains; the (n, γ) reactions add a branch to each radioactive nuclide and couple stable nuclides. It is possible to generate nuclides not present in the absence of the neutron flux. The result is that potentially there are over 250 nuclides which may contribute to the macroscopic absorption cross section of the reactor. The actual number which need be included is dependent on the maximum flux-time of interest, because many of the nuclides do not directly produce or lead to a significant absorption cross section within existing or contemplated reactor lifetimes. The individual nuclide concentrations and, therefore, the total aggregate absorption cross section is a function of time through radioactive decay and of flux-time and spectrum through neutron absorption and fission. For example, consider a particular fission product such as $(36h)Rh-105$ with its $\sim 15,000$ -barn thermal cross section. Its concentration at any time depends on the time integral of its fission source and depletion rate and on its formation rate from other nuclide transmutations. However, its depletion rate depends on the sum of its decay and neutron absorption rates, hence, on the flux level and spectrum, and its formation rate depends on the flux level, which in turn depends on the degree of fuel depletion. The aggregate absorption cross section assumes all of these dependences. Clearly, because of radioactive decay, the aggregate poison may be significantly dependent on the operating history of the reactor, and concomitantly, it may exhibit a significant transient behavior* following power variations (in addition to the well known Xe-135 and Sm-149 transients). Whether or not the aggregate cross section exhibits a significant history dependence or a net transient behavior, its individual nuclide contributions and, hence, the aggregate value can only be accurately calculated by accounting for all of the coupling systematics.

The previous discussion is sufficient to illustrate that if one attempts to describe the fission product group in its entirety by accounting for both radioactive decay and neutron absorption, one appears to face a formidable task. On the other hand because of the long irradiation times and closely designed reactivity control involved with advanced reactor designs, the accumulation of the fission product poison with irradiation time and any transient behavior it exhibits are of increasing importance. It is difficult to estimate the effect of any simplifying calculational assumption because of the multiplicity of factors which enter into the determination of the composition of the fission product group after a considerable irradiation time. Therefore, a complete treatment is highly desirable.

Another major factor enters into any attempt to treat the fission products explicitly. This is the consideration of the state of knowledge of the absorption cross sections and fission yields of the products. This set of physical data is the most important factor which governs the meaningfulness of the result. After examining the available physical data, it was concluded that a thorough analytical treatment of the fission product group was warranted. In addition there exists an experimental program within the PWR Project when the objective is to measure the fission product absorption of irradiated fuels. This program provides a check on the reasonableness of the analytical results.

*"Transient behavior" is used here to refer to changes in poison following any change in power level; i.e., it does not necessarily mean transitory changes.

Insofar as possible, the basic physical description of the fission products has been incorporated into Part I of this report, and Part II is devoted to the numerical results. More specifically, Part I contains:

1. The development of a very general analytical treatment, applicable to all fuels, which includes radioactive decay and neutron capture, where the time-dependent fluxes and spectrum are assumed to be independently known
2. A brief discussion of a FORTRAN program, CINDER (Reference 6) based on the analysis
3. A collection and summary of the physical data (cross sections, half-lives, yield fractions, and nuclide chains) necessary to describe the products resulting from fission of U-235, U-238, and Pu-239

Part II uses the treatment and data of Part I:

1. To determine, under a variety of reactor conditions, the characteristics and sensitivity of the time or depletion dependent aggregate absorption cross section with respect to variations in flux level, spectrum, and flux-time histories
2. To identify the important fission products and nuclide chains
3. To investigate the effect of making simplifying assumptions in the analytical treatment, using the exact treatment as a standard
4. To compare calculations with experimental measurements of the gross fission product poison and, thereby, estimate the adequacy of the presently known physical data
5. To estimate the net uncertainty associated with factors entering into the calculations

The analytical method is exact in its treatment of all coupling systematics; therefore, it represents a considerable departure from previous methods. The treatment is quite general and flexible so that new physical data is readily incorporated. The fission products from the fuels which are both generated (e.g., Pu-239) and depleted (e.g., U-238), and the generated fuel poisons, such as Pu-240, can be calculated. The description of this treatment is, therefore, the most important aspect of this report. In a sense the calculations are secondary; it must be realized that any study of fission product absorption is necessarily of a continuing nature and will remain so for some time, because there are many products for which no cross-sectional information is yet available. However, an attempt was made to make the physical data used in this report as accurate and as complete as possible, and it is therefore worth noting at this point that a detailed comparison with recent experimental measurements indicates agreement within 2 percent thermally and 15 percent epithermally, with the calculation on the high side. This agreement on the aggregate cross section could be due to a fortuitous cancellation of errors among the individual constituents, but it hopefully indicates that there are no remaining fission product poisons of significance, at least after the extensive irradiation times involved in the comparison.

Some reports on fission products include the poison from isotopes formed by neutron capture in fuel, such as U-236; some exclude Xe-135 and, frequently, Sm-149. Therefore, it is appropriate to note here that the numerical results in this report are confined to the poison from fission products only. All known fission products are included in the reported results except the Xe-135 and Sm-149 nuclides generated by the fission yields to isobaric chains of mass 135 and 149. Depletion programs generally account explicitly for these two nuclides. Poison generated by capture in the fuel materials have been removed from all results reported herein.

PART I. ANALYTICAL TREATMENT AND NUCLIDE DATA

I. THEORY

Since the neutron-to-proton ratio in fissile nuclides is larger than the ratio in stable nuclides with intermediate masses, the fission fragments are formed with ratios that are too large for maximum stability. This remains true even after the emission of the prompt and delayed neutrons. Consequently, a description of the fission products must consider mostly β^- -decay and neutron absorption as coupling mechanisms. In a few cases γ -decay from excited isomeric states must also be described. These reactions lead to some complicated branchings which are typified in the schematic representation of some specific nuclides in the chain containing Xe-135:

Half-lives, yield fractions, and cross sections are omitted for clarity.

In principle it would be possible to solve the differential equations which describe all such couplings that actually exist among the fission products, but it appears that such a specific and inflexible description, even if feasible, could not be easily altered if later data indicated the need to add other branches. A more basic approach, leading to a unique set of coupled equations which can be solved independently of particular nuclide parameters and applied to all chains, is obtained by resolving each such chain as (1) above into several sub-chains of nuclides, each nuclide being coupled to a single parent. The exact process is described in Appendix A. Such chains exhibit no branches and will be referred to as linear chains. For example, consider the branch following Xe-135:

This can be resolved into:

and

The direct fission yields of the nuclides can be arbitrarily divided between the nuclides common to chains 2a and 2b. The chains are independent in the sense that the concentrations in each chain can be calculated without regard to the other chain. The total concentration of each nuclide is the sum of the partial concentration from each chain. A complete resolution of chain (1), for the nuclides explicitly noted would require seven linear chains. In some of these the partial concentrations of certain nuclides would be duplicated, and therefore, the partial concentrations must be summed with care to include each concentration only once.

This linear chain resolution would be useless in the analysis of fission products if all nuclides were strongly coupled and each exhibited the general branching shown in Appendix A. However, this is not the case. The short-lived nuclides are, effectively, only β^- -coupled, and the stable nuclides are coupled only by neutron absorption. Even those radioactive nuclides with significant absorption cross sections generally possess only one dominant branch, and, often, one of two equally active branches leads to a stable nuclide which has no significant absorption cross section; the branch need not be followed. Since the differential equations which describe the coupled nuclides are linear, a sufficient condition for such resolutions to be possible is that no nuclide concentration be dependent on the concentration of any of its progeny. This condition is satisfied by the fission products, since the only reactions of importance are (n, γ) , β^- decay, and γ -decay from isomeric states.

This treatment by linear chains involves no loss in generality but results in a considerable increase in flexibility. In terms of a few energy group structure, the concentration of the i^{th} nuclide in a chain is determined by the following sequence of coupled equations:

$$\frac{dN_i}{dt} = \sum_k y_i^k S^k + \gamma_{i-1} N_{i-1} - (\lambda_i + A_i) N_i \quad (1)$$

$$i = 1, 2, 3 \dots$$

where y_i^k = yield fraction from fissile nuclide k

S^k = fission rate of the k^{th} fissile nuclide

λ_i = radioactive decay constant

$A_i = \sum_j g_i^j \sigma_i^j \phi^j$ where σ_i^j is the total (n, γ) absorption cross section of the i^{th} nuclide in energy group j, g_i^j is a spectrum or shielding factor, and ϕ^j is the neutron flux in the j^{th} energy group.

γ_{i-1} = either λ_{i-1} or A_{i-1} , depending on coupling from precursor i-1

(Here, A_{i-1} is not necessarily the total absorption rate, but it is only that portion of the (n, γ) reaction rate leading to N_i ; thus, in fissile nuclides A_{i-1} is the capture rate.)

This set of equations can equally well describe the fissile nuclides and nuclides formed by neutron capture in the fuel chains (in which case $y=0$) or the fission product chains.

The general solution of the set (1) for the n^{th} nuclide, assuming a constant flux and an average fission rate during time increment Δt , is shown in Appendix B to be:

$$N_n(t + \Delta t) = \sum_{m=1}^n \frac{1}{\gamma_n} \frac{n}{\pi} \frac{n}{k-m} \gamma_k \left\{ \overline{y_m S} \left[\frac{1}{\frac{n}{\pi} (\lambda_l + A_l)} \right] - \sum_{j=m}^n \frac{\exp [- (\lambda_j + A_j) \Delta t]}{(\lambda_j + A_j) \frac{n}{\pi} \left(\lambda_i + A_i - \lambda_j - A_j \right)} \right\} + N_m(t) \left(\sum_{j=m}^n \frac{\exp [- (\lambda_j + A_j) \Delta t]}{\frac{n}{\pi} (\lambda_i + A_i - \lambda_j - A_j)} \right) \quad (2)$$

where $\overline{y_m S} = \sum_k y_m^k S^k$ is the time averaged direct yield rate to nuclide m during the interval Δt summed over all fissile nuclides, k.

The physical significance of the various parts of this equation is discussed in Reference (6), a companion report on a computer program. Since Δt can be made arbitrarily small, the use of a constant flux per time increment presents no practical limitation on the use of Equation (2).

It is emphasized that this solution fits all linear chains of any length; hence, it admits of a very general digital program capable of handling any sequence of transmutations involving all forms of radioactive decay and neutron absorption. For this report only fuel and fission product chains are of interest.

Using the concentration from Equation (2), the macroscopic absorption cross section of nuclide n in energy group j is:

$$\Sigma_n^j = g_n^j \sigma_n^j N_n \quad (3)$$

A more convenient quantity is the ratio of Σ_n^j to the total number of fissions which have occurred. This quantity is appropriately referred to as barns/fission (b/f):*

$$\hat{\sigma}_n^j = \frac{\Sigma_n^j}{\sum_k \int_0^t s^k dt} \quad (4)$$

The total barns/fission is then

$$\hat{\sigma}^j = \sum_n \hat{\sigma}_n^j \quad (5)$$

Other quantities are sometimes quoted such as barns-per-initial-fuel-atom or barns-per-fuel-atom-destroyed, but barns/fission is used exclusively in this report. $\hat{\sigma}^j$ is most convenient for use in the Bettis depletion programs (Reference 7).**

II. DIGITAL PROGRAM

A FORTRAN program (Reference 6), hereafter referred to as CINDER, has been built around Equation (2), using one to four arbitrary energy groups. It will handle up to 300 nuclides grouped into linear chains as specified by the requestor. Up to 99 fission product chains and 40 fuel chains containing a maximum of 15 nuclides each are permitted. The direct fission yields for the nuclides in the fission product chains can be specified from up to 10 fissile nuclides appearing in the fuel chains. These limitations are arbitrary and readily changed. The calculation is performed in a sequence of time increments—each of arbitrary length. At each of these increments, the power density (or, optionally, the thermal flux), the fast-to-thermal flux ratios, a thermal spectrum (or shielding) factor, and an epithermal shielding factor (per nuclide) can be specified. CINDER is thus a general, "one-point" program which evaluates the time-dependent concentrations of coupled nuclides following irradiation in a specified time-dependent flux, and it is therefore generally useful outside of fission product calculations. The parameters needed to calculate the concentrations (decay constants, cross sections, type of coupling, yields, etc.) must be specified by the requestor. A recommended set of data is offered in Table 1.

At the end of each timestep the program output provides the individual nuclide concentrations, the barns/fission and macroscopic absorption cross sections in each energy group, and other quantities of interest. A summation of the barns/fission and macroscopic cross sections over selected nuclides in each energy group is also given. This makes it possible to selectively exclude nuclide poisons from the summation which would otherwise be counted more than once because of the linear chain resolutions, as well as any other special nuclide poisons, such as Xe-135 and nuclide poisons in the fuel chains.

The program incorporates a self-check against computer round-off errors, which also helps to reduce running time. For the 67 chains of 256 nuclides used in this analysis, the running time is ~0.0012 minutes per nuclide per timestep on the Philco-2000 computer (2 μ -second memory).

*Throughout this report, the circumflex ($\wedge$) will be used to distinguish the barns-per-fission cross sections from the nuclide microscopic cross sections.

**In the Bettis programs $\hat{\sigma}^j$ is used for element 28 with all $\gamma_{m,A} = 1$ and $\theta_{28} = 0$.

TABLE 1. LINEAR CHAINS AND NUCLIDE PARAMETERS

Chain No.	Nuclide	Coupling††	Half-Life	λ	σ_{220}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
1	Zn-72	λ	49h	3.929×10^{-6}	0	0	1.6×10^{-5}	1.6×10^{-5}	1.6×10^{-5}
	Ga-72	λ	14.1h	1.366×10^{-5}	0	0	0	0	0
	Ge-72	A	∞	0	0.93	0.043	0	0	0
	Ge-73	A	∞	0	14	0.61	1.1×10^{-4}	1.1×10^{-4}	1.1×10^{-4}
	Ge-74	0	∞	0	0.62	0.027	3×10^{-4}	3×10^{-4}	3×10^{-4}
(End of Chain)									
2	Ge-76	0	∞	0	0.36	0.016	0.015	0.015	0.015
3	As-75	0	∞	0	4.3	0.19	8×10^{-4}	8×10^{-4}	8×10^{-4}
4	As-77	λ	58.7h	4.975×10^{-6}	0	0	0.0083	0.0038	0.01
	Se-77	A	∞	0	42	3.15	0	<0>	0
	Se-78	0	∞	0	0.4	0.017	0.02	<0.02>	0.02
5	Se-79	A	6×10^4 y	~ 0	40	1.74	0.056	<0.047>	0.04
	Se-80	0	∞	0	0.61	0.092	0.06	<0.065>	0.07
6	Br-81	0	∞	0	3.3	6.74	0.14	<0.12>	0.11
7	Kr-82	A	∞	0	45	21.55	0.28	<0.22>	0.17
	Kr-83	A	∞	0	205	24.11	0.544	0.40	0.29
	Kr-84	0	∞	0	0.16	0.325	1.00	0.85	0.47
8	Kr-85	λ	10.6y	2.074×10^{-9}	8.0	0.61	0.293	0.153	0.122
	Rb-85	A	∞	0	0.91	0.32	1.007	0.525	0.418
	Sr-86	0	∞	0	1.65	0.057	0	<0>	0
9	Kr-85	A	10.6y	2.074×10^{-9}	8.0	0.61	0.293*	0.153*	0.122*
	Kr-86	0	∞	0	0.06	0.094	2.02	1.38	0.75
10	Rb-87	A	5×10^{10} y	~ 0	0.12	0.112	2.49	<1.7>	0.91
	Sr-88	0	∞	0	0.005	0.30	3.57	<2.50>	1.4

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
11	Sr-89	λ	50.5d	1.589×10^{-7}	0.5	0.0183	4.79	2.9	1.7
	Y-89	A	∞	0	1.31	0.19	0	<0>	0
	Zr-90	0	∞	0	0.10	0.0044	0	<0>	0
12	Sr-90	λ	28y	7.844×10^{-10}	1.0	0.17	5.77	3.2	2.2
	Zr-90	0	∞	0	0.10	0.0044	0	<0>	0
13	Y-91	λ	58d	1.383×10^{-7}	1.07	0.048	5.4	<4.0>	2.55
	Zr-91	0	∞	0	1.58	0.99	0.44	<0.22>	0
14	Zr-92	0	∞	0	0.25	0.30	6.03	<4.56>	3.1
15	Zr-93	0	1.1×10^6 y	~ 0	1.1	3.08	6.45	<5.18>	3.9
16	Zr-94	0	∞	0	0.08	0.29	6.40	<5.45>	4.5
17	Zr-95	λ	65d	1.234×10^{-7}	0	0	6.20	5.7	5.0
	Nb-95	λ	35d	2.292×10^{-7}	0	0	0.07	<0.035>	0
	Mo-95	A	∞	0	13.9	11.61	0	<0>	0
	Mo-96	A	∞	0	1.2	2.91	0	<0>	0
	Mo-97	A	∞	0	2.2	1.76	6.09	<5.84>	5.6
	Mo-98	0	∞	0	0.51	0.61	5.78	<5.84>	5.9
18	Zr-96	0	∞	0	0.053	0.0044	6.33	<5.72>	5.1
19	Mo-99	λ	66h	2.917×10^{-6}	0	0	6.06	6.3	6.1
	Tc-99	A	2.1×10^5 y	~ 0	22	24.6	0	<0>	0
	Ru-100	A	∞	0	7.5	0.33	0	<0>	0
	Ru-101	A	∞	0	5	8.47	5.0	<5.45>	5.9
	Ru-102	A	∞	0	1.44	1.12	4.1	<5.0>	5.9
	Ru-103	λ	39.7d	2.021×10^{-7}	0	0	3.0	6.6	5.6
	Rh-103	A	∞	0	149	113.3	0	<0>	0
	Pd-104	A	∞	0	22.5	2.63	0	<0>	0
	Pd-105	0	∞	0	1.0	7.81	0	<0>	0

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
20	Mo-100	0	∞	0	0.5	0.695	6.30	<6.7>	7.1
21	Ru-104	0	∞	0	0.7	0.71	1.8	<3.85>	5.9
22	Rh-105	λ	36h	5.348×10^{-6}	15,000	653.10	0.9	<3.20>	5.5
	Pd-105	A	∞	0	10	7.81	0	<0>	0
	Pd-106	A	∞	0	6	1.32	0	<0>	0
	Pd-107	A	7×10^6 y	~ 0	10	4.4	0	<0>	0
	Pd-108	A	∞	0	10.4	16.98	0	<0>	0
	Ag-109	0	∞	0	87	158.14	0	<0>	0
23	Rh-105	A	36h	5.348×10^{-6}	15,000	653.10	0.9*	<3.20>*	5.5*
	Pd-106	A	∞	0	6	1.32	0	0	0
	Pd-107	A	7×10^6 y	~ 0	10	4.4	0	0	0
	Pd-108	A	∞	0	10.4	16.98	0	0	0
	Ag-109	0	∞	0	87	158.14	0	0	0
24	Ru-106	A	1.01y	2.175×10^{-8}	9	1.72	0.38	2.7	4.5
	Pd-107	A	7×10^6 y	~ 0	10	4.4	0.19	<1.9>	3.6
	Pd-108	A	∞	0	10.4	16.98	0.09	<1.34>	2.6
	Ag-109	0	∞	0	87	158.14	0.03	0.32	1.55
25	Ru-106	λ	1.01y	2.175×10^{-8}	9	1.72	0.38*	2.7*	4.5*
	Pd-106	A	∞	0	6	1.32	0	<0>	0
	Pd-107	A	7×10^6 y	~ 0	10	4.4	0	<0>	0
	Pd-108	A	∞	0	10.4	16.98	0	<0>	0
	Ag-109	0	∞	0	87	158.14	0	<0>	0
26	Pd-110	0	∞	0	0.21	0.669	0.021	<0.28>	0.55

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
27	Ag-111	λ	7.6d	1.056×10^{-6}	0	0	0.019	0.076	0.20
	Cd-111	A	∞	0	85	5.46	0	<0>	0
	Cd-112	A	∞	0	0.03	1.52	0.01	0.046	0.12
	Cd-113	A	∞	0	20,000	71.73	0.012	<0.041>	0.07
	Cd-114	0	∞	0	1.24	1.153	0.011	<0.026>	0.04
28	Cd-115m	λ	43d	1.866×10^{-7}	0	0	0.0007	0.003	0.003
	In-115	A	6×10^{14} y	~ 0	207	359	0	<0>	0
	Sn-116	0	∞	0	0.006	1.496	0	<0>	0
29	Cd-115	λ	53h	3.633×10^{-6}	0	0	0.00922	0.034	0.030
	In-115	A	6×10^{14} y	~ 0	207	359	0	<0>	0
	Sn-116	0	∞	~ 0	0.006	1.496	0	<0>	0
30	Cd-115	λ	53h	3.633×10^{-6}	0	0	0.00048	0.002	0.002
	Sn-115	A	∞	0	60	2.82	0	<0>	0
	Sn-116	0	∞	0	0.006	1.496	0	<0>	0
31	Cd-116	A	∞	0	1.5	0.13	0.011	<0.023>	0.035
	Sn-117	A	∞	0	2.7	1.768	0.011	<0.023>	0.035
	Sn-118	A	∞	0	0.01	0.479	0.01	<0.023>	0.035
	Sn-119	0	∞	0	2.5	0.725	0.011	<0.023>	0.035
32	Sn-120	0	∞	0	0.141	0.089	0.011	<0.023>	0.035
33	Sn-121	λ	27.5h	7.002×10^{-6}	0	0	0.015	<0.025>	0.035
	Sb-121	A	∞	0	5.9	22.27	0	<0>	0
	Sb-122	λ	2.8d	2.865×10^{-6}	0	0	0	<0>	0
	Te-122	0	∞	0	2.8	6.17	0	<0>	0
34	Sn-122	0	∞	0	0.161	0.007	0.013	<0.024>	0.035

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
35	Sn-123	λ	136d	5.899×10^{-8}	0	0	0.0013	<0.021>	0.04
	Sb-123	A	∞	0	4.1	17.77	0	<0>	0
	Sb-124	λ	60d	1.337×10^{-7}	0	0	0	<0>	0
	Te-124	0	∞	0	6.8	0.527	0.017	<0.028>	0.04
36	Sn-125	λ	9.6d	8.357×10^{-7}	0	0	0.013	<0.032>	0.05
	Sb-125	λ	2.0y	1.098×10^{-8}	1.56	0.068	0.008	<0.019>	0.03
	Te-125	0	∞	0	1.56	2.60	0	<0>	0
37	Te-126	0	∞	0	0.8	1.02	0.10	<0.14>	0.18
38	Sb-127	λ	91h	2.116×10^{-6}	0	0	0.028	0.026	0.045
	Te-127m	λ	105d	7.641×10^{-8}	0	0	0.007	<0>	0
	I-127	0	∞	0	7	17.87	0.113	<0.14>	0.167
39	Sb-127	λ	91h	2.116×10^{-6}	0	0	0.102	0.094	0.158
	I-127	0	∞	0	7	17.87	0	<0>	0
40	Te-128	0	∞	0	0.3	1.13	0.37	<0.56>	0.74
41	Te-129m	λ	37d	2.168×10^{-7}	0	0	0.35	<0.45>	0.544
	I-129	A	$1.7 \times 10^7 \text{ y}$	0	32	4.26	0.45	<0.653>	0.856
	I-130	λ	12.6h	1.528×10^{-5}	18	0.78	0	<0>	0
	Xe-130	0	∞	0	2.5	0.11	0	<0>	0
42	Te-130	0	∞	0	0.5	0.022	2.0	<2.25>	2.5
43	Te-131m	λ	30h	6.418×10^{-6}	0	0	0.44	0.45	0.54
	I-131	λ	8.05d	9.966×10^{-7}	50	2.18	2.66	2.75	3.26
	Xe-131	A	∞	0	120	87.98	0	<0>	0
	Xe-132	0	∞	0	0.2	0.95	0	<0>	0

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
44	Te-132	λ	77h	2.50×10^{-6}	0	0	4.7	4.7	5.3
	Xe-132	0	∞	0	0.2	0.95	0	<0>	0
45	I-133	λ	20.8h	9.257×10^{-6}	0	0	6.9	5.5	6.9
	Xe-133	A	5.27d	1.522×10^{-6}	190	8.27	0	<0>	0
	Xe-134	A	∞	0	0.2	0.3	8.06	6.6	7.5
	Xe-135	0	9.2h	2.093×10^{-5}	2.72×10^6	500	0	0	0
46	I-133	λ	20.8h	9.257×10^{-6}	0	0	6.9*	5.5*	6.9*
	Xe-133	λ	5.27d	1.522×10^{-6}	190	8.27	0*	<0>*	0*
	Cs-133	A	∞	0	29	46.21	0	<0>	0
	Cs-134	λ	2.3y	9.556×10^{-9}	134	6.10	0	<0>	0
	Ba-134	0	∞	0	2	0.087	0	<0>	0
47	I-133	λ	20.8h	9.257×10^{-6}	0	0	6.9*	5.5*	6.9*
	Xe-133	λ	5.27d	1.522×10^{-6}	190	8.27	0*	<0>*	0*
	Cs-133	A	∞	0	29	46.21	0*	<0>*	0*
	Cs-134	A	2.3y	9.550×10^{-9}	134	6.10	0*	<0>*	0*
	Cs-135	0	$2.6 \times 10^6 \text{ y}$	~ 0	8.7	6.81	0	<0>	0
48	I-135	λ	6.7h	2.874×10^{-5}	0	0	6.1**	5.7**	6.85**
	Xe-135	λ	9.2h	2.093×10^{-5}	2.72×10^6	500	0.2**	0.18**	0.22**
	Cs-135	A	$2.6 \times 10^6 \text{ y}$	~ 0	8.7	6.81	0.11	0.12	0.13
	Cs-136	λ	13d	6.171×10^{-7}	0	0	0	<0>	0
	Ba-136	0	∞	0	0.4	1.45	0	<0>	0
49	I-135	λ	6.7h	2.874×10^{-5}	0	0	6.1*	5.7*	6.85*
	Xe-135	A	9.2h	2.093×10^{-5}	2.72×10^6	500	0.2*	0.18*	0.22*
	Xe-136	0	∞	0	0.15	0.053	6.46	5.9	6.6

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
50	Cs-137	λ	30y	7.327×10^{-10}	0.11	1.28	6.15	6.2	6.5
	Ba-137	0	∞	0	5.1	0.22	0	<0>	0
51	Ba-138	A	∞	0	0.7	0.088	5.74	<6.0>	6.3
	La-139	A	∞	0	8.9	1.21	6.55	<6.28>	6.0
	La-140	λ	40.2h	4.79×10^{-6}	3.1	0.135	0	<0>	0
	Ce-140	0	∞	0	0.66	0.086	0	<0>	0
52	Ba-140	λ	12.8d	6.263×10^{-7}	12	0.522	6.44	5.7	5.6
	La-140	λ	40.2h	4.79×10^{-6}	3.1	0.135	0	<0>	0
	Ce-140	0	∞	0	0.66	0.083	0	<0>	0
53	Ce-141	λ	33d	2.431×10^{-7}	0	0	6.0	<5.8>	5.6
	Pr-141	A	∞	0	11.6	2.59	0	<0>	0
	Pr-142	λ	19.3h	9.976×10^{-6}	18	0.784	0	<0>	0
	Nd-142	A	∞	0	18	0.784	0	<0>	0
	Nd-143	A	∞	0	324	16.13	0	<0>	0
	Nd-144	0	5×10^{15} y	~ 0	5	1.11	0	<0>	0
54	Ce-142	0	∞	0	1	0.103	6.01	<5.50>	5.0
55	Ce-143	λ	33h	5.835×10^{-6}	6	0.261	6.0	<5.19>	4.38
	Pr-143	λ	13.8d	5.813×10^{-7}	89	20.91	0.03	<0.125>	0.22
	Nd-143	A	∞	0	324	16.13	0	<0>	0
	Nd-144	A	5×10^{15} y	~ 0	5	1.11	0	<0>	0
	Nd-145	0	∞	0	60	26.93	3.98	<3.54>	3.1

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
56	Nd-145	A	∞	0	60	26.96	3.98*	<3.54>*	3.1*
	Nd-146	A	∞	0	10	1.22	3.07	<2.84>	2.6
	Nd-147	λ	11.1d	7.228×10^{-7}	0	0	2.36	2.6	2.05
	Pm-147	C†	2.6y	8.454×10^{-9}	232†	359.8†	0	<0>	0
	Pm-148m	A	40.6d	1.9756×10^{-7}	27,000	3,521	0	<0>	0
	Pm-149	λ	53.1h	3.626×10^{-6}	0	0	0	<0>	0
	Sm-149	A	∞	0	40,800	374.1	0	<0>	0
	Sm-150	A	∞	0	85	50.61	0	<0>	0
	Sm-151	A	80y	2.746×10^{-10}	15,400	275.1	0	<0>	0
	Sm-152	A	∞	0	216	261	0	<0>	0
	Sm-153	λ	47h	4.097×10^{-6}	0	0	0	<0>	0
	Eu-153	A	∞	0	450	151.8	0	<0>	0
	Eu-154	A	16y	1.373×10^{-9}	1,500	65.31	0	<0>	0
	Eu-155	0	4y	5.495×10^{-9}	14,000	610	0	<0>	0
57	Nd-145	A	∞	0	60	26.96	3.98*	<3.54>*	3.1*
	Nd-146	A	∞	0	10	1.22	3.07*	<2.84>*	2.6*
	Nd-147	λ	11.1d	7.228×10^{-7}	0	0	2.36*	2.6*	2.05*
	Pm-147	C†	2.6y	8.454×10^{-9}	232	359.80	0*	<0>*	0*
	Pm-148	A	5.39d	1.4884×10^{-6}	1,500	4,841.34	0	<0>	0
	Pm-149	λ	53.1h	3.626×10^{-6}	0	0	0	<0>	0
	Sm-149	A	∞	0	40,800	374.1	0	<0>	0
	Sm-150	A	∞	0	85	50.61	0	<0>	0
	Sm-151	A	80y	2.746×10^{-10}	15,400	275.1	0	<0>	0
	Sm-152	A	∞	0	216	261	0	<0>	0
	Sm-153	λ	47h	4.097×10^{-6}	0	0	0	<0>	0
	Eu-153	A	∞	0	450	151.8	0	<0>	0
	Eu-154	A	16y	1.373×10^{-9}	1,500	65.31	0	<0>	0
	Eu-155	0	4y	5.495×10^{-9}	14,000	610	0	<0>	0

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EFI}	% Yields		
							U-235	U-238	Pu-239
58	Ce-144	λ	280d	2.865×10^{-8}	0.96	0.29	5.62	<4.5>	3.8
	Nd-144	A	5×10^{15} y	~ 0	5	1.11	0	<0>	0
	Nd-145	0	∞	0	60	26.96	0	<0>	0
59	Ce-144	A	280d	2.865×10^{-8}	0.96	0.29	5.62*	<4.5>*	3.8*
	Nd-145	0	∞	0	60	26.96	0	<0>	0
60	Nd-146	A	∞	0	10	1.22	3.07*	<2.84>*	2.6*
	Nd-147	λ	11.1d	7.228×10^{-7}	0	0	2.36*	2.6*	2.05*
	Pm-147	C†	2.6y	8.454×10^{-9}	232	359.80	0*	<0>*	0*
	Pm-148m	λ	40.6c	1.9756×10^{-7}	27,000	3,521	0*	<0>*	0*
	Sm-148	A	∞	0	9	5.50	0	<0>	0
	Sm-149	A	∞	0	40,800	374.1	0	<0>	0
	Sm-150	A	∞	0	85	50.31	0	<0>	0
	Sm-151	0	30y	2.743×10^{-10}	15,400	275.1	0	<0>	0
61	Nd-146	A	∞	0	10	1.22	3.07*	<2.84>*	2.6*
	Nd-147	λ	11.1d	7.228×10^{-7}	0	0	2.36*	2.6*	2.05*
	Pm-147	C†	2.6y	8.454×10^{-9}	232	359.80	0*	<0>*	0*
	Pm-148	λ	5.39d	1.4884×10^{-6}	1,500	4,841.34	0*	<0>*	0*
	Sm-148	A	∞	0	9	5.50	0	<0>	0
	Sm-149	A	∞	0	40,800	374.1	0	<0>	0
	Sm-150	A	∞	0	85	50.61	0	<0>	0
	Sm-151	0	30d	2.746×10^{-10}	15,400	275.1	0	<0>	0
62	Nd-146	A	∞	0	10	1.22	3.07*	<2.84>*	2.6*
	Nd-147	λ	11.1d	7.228×10^{-7}	0	0	2.36*	2.6*	2.05*
	Pm-147	λ	2.6y	8.454×10^{-9}	232	359.80	0*	<0>*	0*
	Sm-147	A	1.3×10^{11} y	~ 0	87	77.87	0	<0>	0
	Sm-148	A	∞	0	9	5.50	0	<0>	0
	Sm-149	A	∞	0	40,800	374.1	0	<0>	0
	Sm-150	0	∞	0	85	50.61	0	<0>	0

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
63	Nd-148	A	∞	0	3.4	5.28	1.71	<1.7>	1.7
	Pm-149	λ	53.1h	3.626×10^{-6}	0	0	1.13**	1.8**	1.3**
	Sm-149	A	∞	0	40,800	374.1	0**	<0>**	0**
	Sm-150	A	∞	0	85	50.61	0	<0>	0
	Sm-151	A	80y	2.746×10^{-10}	15,400	275.1	0	<0>	0
	Sm-152	A	∞	0	216	261	0	<0>	0
	Sm-153	λ	47h	4.097×10^{-6}	0	0	0	<0>	0
	Eu-153	A	∞	0	450	151.8	0	<0>	0
	Eu-154	A	16y	1.373×10^{-9}	1,500	65.31	0	<0>	0
	Eu-155	0	4y	5.495×10^{-9}	14,000	610	0	<0>	0
64	Nd-150	A	∞	0	3	0.13	0.67	<0.84>	1.0
	Pm-151	λ	28.4h	6.780×10^{-6}	0	0	0.44	<0.62>	0.80
	Sm-151	A	80y	2.746×10^{-10}	15,400	275.1	0	<0>	0
	Sm-152	A	∞	0	216	261	0.281	<0.44>	0.60
	Sm-153	λ	47h	4.097×10^{-6}	0	0	0.15	<0.41>	0.34
	Eu-153	A	∞	0	450	151.8	0.019	<0.01>	0
	Eu-154	A	16y	1.373×10^{-9}	1,500	65.31	0	<0>	0
	Eu-155	λ	4y	5.495×10^{-9}	14,000	610	0	<0>	0
	Gd-155	0	∞	0	58,000	179.34	0	<0>	0
65	Sm-154	A	∞	0	5.5	2.72	0.077	<0.16>	0.25
	Eu-155	λ	4y	5.495×10^{-9}	14,000	610	0.033	<0.10>	0.16
	Gd-155	0	∞	0	58,000	179.34	0	<0>	0
66	Eu-156	λ	15.4d	5.209×10^{-7}	0	0	0.014	0.071	0.10
	Gd-156	A	∞	0	7	0.305	0	<0>	0
	Gd-157	0	∞	0	242,000	570	0	<0>	0

TABLE 1 (Cont)

Chain No.	Nuclide	Coupling	Half-Life	λ	σ_{2200}	σ_{EPI}	% Yields		
							U-235	U-238	Pu-239
67	Eu-157	λ	15.4h	1.250×10^{-5}	0	0	0.0078	<0.038>	0.07
	Gd-157	A	∞	0	242,000	570	0	<0>	0
	Gd-158	A	∞	0	3.9	0.174	0.002	<0.018>	0.035
	Gd-159	λ	18h	1.070×10^{-5}	0	0	0.00107	0.0084	0.015
	Tb-159	A	∞	0	46	42.71	0	<0>	0
	Tb-160	λ	73d	1.099×10^{-7}	525	22.86	0	<0>	0
	Dy-160	A	∞	0	100	4.35	0	<0>	0
	Dy-161	A	∞	0	590	130.22	7.6×10^{-5}	<0.002>	0.0039
	Dy-162	A	∞	0	260	294.10	0	0	0
	Dy-163	A	∞	0	380	151.88	0	0	0
	Dy-164	0	∞	0	2,800	121.91	0	0	0

*Identifies those nuclide yields (zero or otherwise) which are accounted for in previous linear chains; hence, the associated nuclide poison is not included in the aggregate poison.

**These nuclide poisons (X-135 and Sm-149) have been excluded from the aggregate poison since they are normally accounted for in depletion codes.

† See the diagram of the Neodymium chains in Part I, Section IV for the reaction cross sections of Pm-147 leading to the two Pm-148 isomers.

†† λ : Indicates radioactive coupling (primarily β^- decay).

A: Indicates coupling by (n, γ) reaction in total absorption cross section.

C: Indicates coupling by (n, γ) reaction in that portion of the absorption cross section leading to its following daughter nuclide. (In this table such capture cross sections were used only for Pm-147. The remaining chains, containing isomers, used a division of yields according to the fraction of the nuclide transformation leading to the daughter isomers since the respective reaction cross sections were not known.)

III. FORMULATION OF CHAINS

Apart from the available physical data discussed in the next section, there are some general considerations which will determine in a practical sense the initiation and termination of each fission product chain. As discussed in the Introduction, each chain will usually begin with a radioactive nuclide and proceed toward a more stable nuclide (taking a gross viewpoint) through successive transformations by radioactive decay and neutron absorptions. The stable and near stable nuclides will be coupled essentially only by the (n, γ) reaction. Therefore, the initiation of each chain will be determined by the shortest lived nuclide which is to be considered, and the chain termination will be determined by the smallest nuclide cross section to be considered which is determined by the maximum energy release or the flux-time of interest. This section outlines the basis for the particular choices used in formulating the chains tabulated in this report.

All nuclides in each isobaric chain with half-lives less than approximately ten hours were assumed to decay instantaneously to the first nuclide having a half-life of ten hours or greater (mass 135 chain excepted). The yield fractions were then appropriately adjusted. Presently available cross sectional data indicates that any transient behavior of the aggregate poison associated with nuclides having smaller half-lives would be minimal in comparison with the Xe-135 transient. Cross sections of the short-lived nuclides are generally unknown, but the cross sections would have to be very large in order for the nuclides to produce a significant poison or for there to be any significant branching due to neutron absorption.

In order to make these statements more quantitative, consider the case of a constant flux and fission rate and a single fuel source. It is convenient to define an effective cross section, $\bar{\sigma}$:

$$\bar{\sigma} = g^4 \sigma^4 + g^3 \sigma^3 \phi^3 / \phi^4, \quad (6)$$

where 4 and 3 denote the thermal and epithermal energy groups respectively. Similarly, an effective barns/fission, $\frac{\Lambda}{\bar{\sigma}}$, can be defined using $\hat{\sigma}$ (Equation 4) from each energy group (dropping the subscript, n):

$$\frac{\Lambda}{\bar{\sigma}} = \frac{\Lambda^4}{\sigma^4} + \frac{\Lambda^3}{\sigma^3} \phi^3 / \phi^4. \quad (7)$$

Then, considering the nuclide of interest to be the first chain nuclide, its concentration, $N(t)$, from Equation (2) is:

$$N(t) = \frac{yS \left[1 - e^{-(\lambda + \bar{\sigma} \phi^4)t} \right]}{\lambda + \bar{\sigma} \phi^4}, \quad (8)$$

and

$$\frac{\Lambda}{\bar{\sigma}}(t) = \frac{y \frac{\Lambda}{\bar{\sigma}} \left[1 - e^{-(\lambda + \bar{\sigma} \phi^4)t} \right]}{(\lambda + \bar{\sigma} \phi^4)t}. \quad (9)$$

At large t the number density approaches an equilibrium, or saturation value, and $\frac{\Lambda}{\bar{\sigma}}$ asymptotically approaches a $1/t$ behavior:

$$N = \frac{yS}{\lambda + \bar{\sigma} \phi^4}; \quad (10)$$

$$\hat{\bar{\sigma}}(t) = \frac{y\bar{\sigma}}{(\lambda + \bar{\sigma}\phi^4)t} \quad (11)$$

Assuming the values:

$$t = 1000 \text{ hours}$$

$$\phi^4 = 10^{14} \text{ cm}^{-2}\text{-sec}^{-1}$$

$$\lambda = 3.85 \times 10^{-5} \text{ sec}^{-1} \text{ (five hour half-life)}$$

$$y = 0.06 \text{ (near the maximum of the individual fission product yields)}$$

then, in order for $\hat{\bar{\sigma}}$ to be one barn (which is <1 percent of the aggregate value), $\bar{\sigma}$ must be ~2000 barns. For a shorter half-life and/or a longer irradiation time the effective microscopic cross section would have to be correspondingly larger. (Almost all of the neglected nuclides had half-lives much smaller than ten hours.) Thus, for a one hour half-life and $t = 10,000$ hours $\bar{\sigma}$ must be >100,000 barns for $\hat{\bar{\sigma}}$ to be one barn. In order for 10% of the daughters of a five-hour nuclide to be formed by neutron capture, its effective cross section must be >40,000 barns.

The over-all transient behavior of a nuclide chain associated with a radioactive nuclide depends on the cross sections of its progeny. However, transient changes in the total chain value of $\hat{\bar{\sigma}}$, caused by a radioactive nuclide, can be no more significant than the $\hat{\bar{\sigma}}$ of the radioactive nuclide unless its progeny possess larger microscopic cross sections. Therefore, the transients associated with the very short-lived nuclides are expected to be no more significant than their poison during steady-state power and, therefore, negligible. (There is some elaboration on these points in Part II, Section V.)

All things considered, there appears to be no justification at this time for explicitly retaining nuclides with half-lives less than about ten hours, not only because of the paucity of cross sectional data for short half-lived nuclides but also because of the unlikelihood of significant poisons.

As will be seen, some of the radioactive nuclides with half-lives >10 hours which are explicitly retained in this analysis have no measured cross sections. They significantly delay the buildup of their more stable progeny. In addition, their half-lives are generally long enough for each to attain a sizeable concentration and, hence, to have a significant effect on the transient behavior of their respective chains, while in most cases their half-lives are short enough that any cross section they possess is not likely to be large enough to significantly affect their calculated concentrations. A familiar case in point is the Sm-149 transient due to its parent (53.1h) Pm-149, which has no measured cross section. Unless the Pm-149 cross section is several thousand barns, it will not invalidate the transients which reactor designers calculate due to its decay into the 40,800-barn Sm-149 cross section. There is, of course, some effect (plus or minus) on the total poison resulting from such radioactive nuclides, and this introduces an uncertainty in the calculated poison, particularly at small fuel depletions and in the transient behavior. In regard to small depletions the limit of Equation (9) for nuclide n at zero depletion is:

$$\hat{\bar{\sigma}}_n(0) = y_n \bar{\sigma}_n, \quad (12)$$

and the aggregate value is

$$\hat{\bar{\sigma}} = \sum_n y_n \bar{\sigma}_n. \quad (13)$$

That is, only the nuclides with direct fission yields contribute to $\frac{\Delta}{\sigma}$. Because most of the direct fission yields go to the very short-lived nuclides, their cross sections really determine the value of $\frac{\Delta}{\sigma}(t)$ at small fuel depletions. Therefore, because of the lack of cross sectional data of the short-lived nuclides, $\frac{\Delta}{\sigma}$ cannot be meaningfully calculated below a fuel depletion of roughly 4 percent. However, for such a small depletion the macroscopic fission product poison is near zero and is therefore of no practical interest.

In this report the initiation of a linear chain begins with the first nuclide having a half-life of ≥ 10 hours. All subsequent known branching is followed until a stable nuclide is reached for each branch which has an effective cross section on the order of one barn. The termination at this point requires some explanation.

Consider the case of two coupled, stable nuclides, again using effective cross sections defined by Equations (6 and 7). $\frac{\Delta}{\sigma}$ for the first nuclide due to its direct yield is given by Equation (9), which can be written more conveniently in the form:

$$\frac{\Delta}{\sigma}_1 = \frac{y_1 \bar{\sigma}_1 \left[1 - e^{-\alpha_1 t} \right]}{\alpha_1 t}, \quad (14)$$

where $\alpha = \lambda + \bar{\sigma} \phi^4$ (for this discussion $\lambda = 0$). Similarly, the barns/fission of the daughter nuclide is:

$$\frac{\Delta}{\sigma}_2 = \frac{y_2 \bar{\sigma}_2}{\alpha_2 t} \left[1 - e^{-\alpha_2 t} \right] + \frac{\alpha_1 y_1 \bar{\sigma}_2}{t} \left\{ \frac{1}{\alpha_1 \alpha_2} - \frac{e^{-\alpha_1 t}}{\alpha_1 (\alpha_2 - \alpha_1)} + \frac{e^{-\alpha_2 t}}{\alpha_2 (\alpha_2 - \alpha_1)} \right\} \quad (15)$$

The last bracketed terms represent the contribution due to the coupling from the first nuclide.

If α_1 is sufficiently small, then

$$e^{-\alpha_1 t} = 1 - \alpha_1 t + O\left(\frac{\alpha_1^2 t^2}{2}\right).$$

This approximation, $1 - \alpha_1 t$, will be accurate to 1 percent for a flux-time of $14 \times 10^{21} \text{ cm}^{-2}$ when

$$\bar{\sigma}_1 \leq \frac{0.02}{\phi t} \cong 1.43 \text{ barns}.$$

To the same order of approximation, the contribution of the first to the second nuclide is zero if the second nuclide also has a cross section on the order of one barn. In short, for two coupled, stable nuclides of sufficiently small cross section there is no significant production of one from the other, and the linear chain may be terminated. However, it is not necessary to follow all chains until such a small cross section is reached. When a nuclide concentration depends on the buildup of a succession of intermediate parents having no direct fission yield and moderate cross sections, then a considerable irradiation time or flux-time is needed for its poison to become significant. The chains then can be terminated with a nuclide possessing a fairly large cross section. However, in the case of successively increasing cross sections in a chain, all such nuclides must be retained, because they all tend to buildup in secular equilibrium with the first nuclide in the chain. The termination, therefore, requires some study for each chain, and it depends on the maximum flux-time for which the chains are to be used. It is believed that, for flux ratios on the order of 10 or less, the chains used in this report are sufficiently long that there will be no more than 1 percent error in the aggregate poison due to chain termination for flux-times up to $\sim 10^{22} \text{ cm}^{-2}$. This represents several seed lives of PWR Core 2. New data may require the addition of more linear chains.

Parenthetically, it is interesting to note from Equation (15) that one can readily deduce that the effective poison of a daughter nuclide due to the parent source can equal but cannot exceed the effective poison of the parent. Such daughters are sometimes referred to as second order products; this label is misleading since it connotes a poison which is second order in magnitude relative to that of the parent, but the terminology is common and will be used in this report.

IV. PHYSICAL DATA

Sixty-seven chains and 256 nuclides are listed in Table 1. The more important nuclides and chains are discussed in Part II, Section II. Because of the resolutions many nuclides appear in more than one chain. There are actually 163 different nuclides listed. The type of coupling, half-life, decay constant, thermal and epithermal cross sections, and yield percent for U-235, U-238, and Pu-239 fissions are given. The chains can be used for other fissile nuclides with appropriate yield changes. When a particular yield is repeated (due to the linear chain resolution), it is indicated by a single asterisk, and the associated nuclide poison is not included in the aggregate summation because it is already accounted for in a previous chain. As previously noted, the Xe-135 and Sm-149 poisons are also excluded in the two chains (48 and 63) which would duplicate the poison which is normally calculated by depletion codes.

The epithermal cross section is defined as:

$$\sigma_{\text{epi}} = \frac{\int_{0.625}^{\infty} \frac{\sigma(E)dE}{E}}{\int_{0.625}^{5530} \frac{dE}{E}} = \frac{RI}{9.0884} \quad (16)$$

Thus, all resonance absorption is assumed to occur in the energy range from 0.625 to 5530 ev where the average cross section is based on a $1/E$ flux. Note that the very small absorption occurring above 5530 ev is approximately accounted for as an absorption in the epithermal energy range. (Correcting the calculated results to other energy divisions than 0.625 ev is straightforward, as shown in Part II, Section IV.) The epithermal cross section was used in the calculations and is therefore listed in Table 1 in preference to the resonance integral. However, these differ only by the constant 9.0884, and all final results will be quoted as resonance integrals.

The resonance integrals were taken from several sources. Most were derived from Walker's comprehensive listing (Reference 2). Many of the more important integrals were taken from the more recent report by Garrison and Roos (Reference 5). When resonance integrals were unavailable from these two compilations, they were taken from the Argonne listing (Reference 8) or computed from the resonance parameters in BNL-325 (Reference 9) with an estimate of the unresolved resonances. When there were no resonance data available for a nuclide having a small thermal cross section, the 2200 m/s cross section was extrapolated with a $1/v$ variation:

$$\sigma_{\text{epi}} = \frac{\sigma_0 \sqrt{.0253} \int_{0.625}^{\infty} \frac{dE}{E^{3/2}}}{\int_{0.625}^{5530} \frac{dE}{E}} = 0.04354 \sigma_{2200} \quad (17)$$

In such cases this would likely be only a lower limit to the actual epithermal cross section. In the few remaining cases where no resonance data are known and the thermal cross section is large, indicating a possible thermal or negative energy resonance, this approximation was not used because it would likely be a gross overestimate of the cross section. Walker's reduced resonance integrals did not include the $1/v$ contribution; therefore, when there were no known resonances below ~ 4 ev, the $1/v$ component was added to the cross sections determined from his data.

The thermal cross sections were obtained almost exclusively from BNL-325 (Reference 9), but a few were taken from the above sources.

The linear chains are based on Katcoff's extensive schematic listing (Reference 10) of the isobaric chains produced by U-235 fission. That is, neutron coupling between the isobaric chains is inferred from the known cross sections, and these additional branches, in addition to those in Katcoff's list, are followed by the linear chain resolution. Nuclide half-lives, U-235 and U-238 yields were also taken from Katcoff's compilation, but the Pu-239 yields were taken from Walker's report. Unfortunately, relatively few U-238 yields have been measured. Those which are known generally fall between the Pu-239 and U-235 yields. Therefore, for expediency an average of the U-235 and Pu-239 yields was used when the U-238 field was unknown. In such cases the average is identified in Table 1 by enclosure in brackets, $\langle y \rangle$.^{*†}

The thermal cross sections and resonance integrals of the very important nuclides Pm-147, (5.39d) Pm-148, Pm-148 and (40.6d) Pm-148m were obtained from Schuman, et al. (Reference 11). These cross sections have been incorporated into chains 56 through 60 of Table 1. They are fed by the 2.36 percent yield to (11.1d) Nd-147. Potentially, these nuclides lead to a production of Sm-149 which can be twice as large as that derived from the direct yield (1.13 percent) of mass chain 149; the latter will be referred to as the normal Sm-149 chain. Therefore, the associated chains are of sufficient importance to deserve elaboration, particularly since they are complicated and not presently well known. The couplings are schematically represented below.

*Walker has recently issued a report containing a compilation of U-238 yields (Reference 15). His recommended values increase the aggregate fission product resonance and thermal cross sections from natural uranium, reported in Part II of this report, by no more than 0.5 and 1.0%, respectively.

†Pu-241 yields are unknown. If it is assumed that its average barns/fission is equal to that of natural U, then the results in Part II need no correction for Pu-241 products.

The small β^- -decay rate of Pm-147 also leads ultimately to Sm-149, but it is not shown here.

The linear chains of particular importance are:

These appear in chains 56 and 57, respectively (Table 1). The remaining branches are chains 58, 59, and 60. They will be collectively referred to as the neodymium chains. Assuming that these cross sections are correct and that the effective Nd-147 cross section is less than about 2000b and the Pm-149 cross section is less than ~5000b, then the following points can be made:

1. Because of the increasing sequence of cross sections and short half-lives from Pm-147 to Sm-149, all nuclides tend to be in secular equilibrium with the buildup of Pm-147 during constant power operation.
2. Because of the relatively small Pm-147 cross sections, this nuclide and those following it do not attain a saturation concentration before a 10 to 20-percent fuel depletion in a power reactor. The actual equilibrium time depends primarily on the magnitude of the epithermal flux because of the relatively large epithermal cross section of Pm-147. (It is perhaps three times the average thermal cross section in a hard spectrum.)
3. Because of the competition between neutron absorption and β^- -decay at the Pm-148 isomers, the net poison is strongly dependent on the flux level and spectrum, since this will determine the rate of Sm-149 production.
4. The equilibrium value of (53.1h) Pm-149 will lead to a large increase in the Sm-149 poison following a reduction to zero power. (This can be 10 to 60 percent of the aggregate poison of the remaining fission products in power reactors.)

Additional comments on this chain and some numerics are in Part II.

PART II. NUMERICAL RESULTS AND RELATED TOPICS

In Part I the basic theory, method of treatment, and physical data were presented. The physical data listed in Table 1 can be used with the CINDER program (Reference 6) to calculate the fission product poison in any specified flux for U-235, U-238, and Pu-239 fuels (or any other fissile nuclides with known yield fractions). This part illustrates the type of results that can be obtained with the digital program together with some general conclusions that can be inferred.

Since the reported calculations are considered to be the first which account for all transformations among the fission products, a wide range of detailed results is presented for U-235, Pu-239, natural, and enriched uranium fuels. The emphasis, however, is on U-235 products. It will be seen that the poison is strongly dependent on fuel depletion or flux-time and has a lesser dependence on flux level, fast-to-thermal flux ratios, thermal spectrum hardness, etc; the results of calculations which isolate and bring out these dependencies are presented. There are no unique values which are applicable to all power reactors, except insofar as one is willing to accept a margin of error and a range of validity, but these are matters of individual need since they depend on the accuracy sought in design calculations. Therefore, all results are best presented in detail rather than as a table of values which approximately apply to a certain class of reactors. However, for U-235 fuel the dependence of the poison on reactor parameters is brought out by simple variations (i.e., changes in spectrum, etc.) about a base case (Figure 1) so that the cross sections applicable to a wide range of core parameters, differing greatly from those of Figure 1, are readily deduced. Thus the results are presented in a manner which simultaneously bring out the characteristics of the fission product cross sections and which will be useful in the inference of poison for the design of a variety of power reactors. The survey is probably not broad enough to satisfy all those readers who desire absorption cross sections for particular reactor designs. The CINDER program is available for the calculation of cross sections which cannot be inferred from the presented survey.*

In addition to the aggregate calculations, this part of the report discusses some related topics including: (1) identification of the important nuclides and a simplified set of chains, (2) effect of radioactive decay on the fission product poison, (3) comparison of calculational and experimental results, and (4) uncertainties in the nuclide data and its effect on the aggregate poison.

I. NUMERICAL RESULTS

All calculations were performed within the following commonly used energy group scheme:

FOUR-GROUP STRUCTURE

<u>Group</u>	<u>Energy Range, e. v.</u>	<u>U (Lethargy)</u>
1	$8.21 \times 10^5 \longrightarrow 10^7$	2.5
2	$5.53 \times 10^3 \longrightarrow 8.21 \times 10^5$	5
3 (epithermal)	$0.625 \longrightarrow 5.53 \times 10^3$	9.0884
4 (thermal)	$0 \longrightarrow 0.625$	--

*Copies of the program may be obtained from: TUG Executive Secretary, Philco Corporation, Computer Division, 515 Pennsylvania Avenue, Fort Washington, Pa.

The fission product cross sections were restricted to groups three and four (Table 1) because, in general, only a thermal cross section and a resonance integral is known for each nuclide. The higher energy groups are needed in order to calculate the behavior of the fuel chains, especially the natural and slightly enriched uranium fuels.

The results of all calculations have been converted to an equivalent 2200 m/s barns/fission (σ_0) and a resonance-integral-per-fission (RI) as follows. From Equations (3 and 4) the barns/fission for each nuclide, n, in each energy group, j, is*

$$\hat{\sigma}_n^j(t) = \frac{g_n^j \sigma_n^j N_n(t)}{\sum_k \int_0^t s^k dt} \quad (18)$$

With the exception of Sm-149, the thermal cross sections of all fission products were assumed to be 1/v. Therefore, each σ_n^4 was conveniently taken to be the 2200 m/s cross section, and all g_n^4 were set equal to a 2200 m/s, 1/v cross section of one barn averaged over the thermal spectrum appropriate to each of the cases considered. Wigner-Wilkins spectra, generated by the KATE program (Reference 12), were used in the averaging. To the extent that the fission products are 1/v, the quantity

$$\sigma_0 = \frac{1}{g^4} \sum_n \hat{\sigma}_n^4 \quad (19)$$

represents an aggregate 2200 m/s cross section per fission. Sm-149 and the fuel cross sections are not 1/v, and the indiscriminate use of g^4 would make a large error in their group-averaged cross sections. Therefore, the correct spectrum-averaged fuel and Sm-149 cross sections were obtained from KATE and appropriately adjusted before insertion into CINDER. The assumption that the remaining fission product cross sections are 1/v is discussed in Section V.

The g-factor for group three would generally be taken as an epithermal self- or mutual-shielding factor. However, as discussed in Section V, no significant shielding was indicated for the calculations reported here so that all g_n^3 were set equal to unity. Therefore, the aggregate value of $\hat{\sigma}^3$ was simply multiplied by the lethargy width of the epithermal group to obtain an aggregate resonance integral-per-fission:

$$RI = 9.0884 \hat{\sigma}^3 \quad (20)$$

The values of σ_0 and RI for all calculations have been plotted versus time, fraction of fuel remaining (α), or megawatt-days per metric ton of uranium (MWD/TONNE). The latter is used for all natural and slightly enriched uranium cases. One comparison of U-235 and Pu-239 poisons has been plotted versus the number of fissions/cm³. α is used for all U-235 fuel cases involving no zero power intervals; otherwise, time is used. The cases showing transients were restricted to variations from constant power operation to zero power.

*The circumflex ($\wedge$) is used to distinguish the barns/fission cross section (except for σ_0) from the microscopic absorption cross section of the nuclides. The bar ($\bar{}$) has been consistently used to denote the effective value (Equations 6 and 7) of either of these quantities.

The parameters noted on all figures are:

N_0 = initial fuel number densities, atoms/cm³

P = power density, watts/cm³

$B_i = \phi^i / \phi^4$, fast-to-thermal flux ratios, $i = 1, 2, 3$

g^4 = thermal spectrum factor: one barn, 2200 m/s, $1/v$ cross section averaged over Wigner-Wilkins spectra

When P or B_i change during depletion, the beginning and end values are noted. None of the calculations used a time-dependent g -factor.

The reader is reminded that the high mass nuclide poisons (U-236, Pu-240, etc.) generated by neutron absorption in fuel, being non-fission products, are not included in the results as sometimes done in fission product studies. Also, the Xe-135 and Sm-149 poisons arising from the direct yields to the mass 135 and 149 isobaric chains were excluded from the aggregate values reported herein. These very important fission product poisons are already explicitly calculated in the Bettis depletion programs (Reference 7), and their magnitude and behavior are well known to all nuclear designers. The Sm-149 poison generated by the neodymium chains described in Part I, Section IV has been included in σ_0 and RI. To be more specific, the only fission product poisons not included in the aggregate value are those of Xe-135 and Sm-149 in chains 48 and 63, respectively, (Table 1).

Most of the following results exhibiting the dependence of the cross sections on the fast-to-thermal flux ratios, thermal spectrum hardness, flux magnitude, power density, and the transient characteristics, are based on U-235 fission. In this way the essential poison characteristics of the products are not as obscured as they would be for natural and slightly enriched uranium fuel caused by the change in average yields following spectrum variations and degree of irradiation.

Figure 1. Reference Case: σ_0 and RI versus Fuel Fraction; Constant Power and Flux Ratio B_3 ; U-235 Fuel.

All variations can be referred to the base case of Figure 1, and it is presumed that the change in poison resulting from two or more parameter variations can be added to obtain the combined effect.

The following comments outline the basis of each figure and offer some qualitative explanation of its characteristics.

Figures 1 through 12 deal exclusively with U-235 fuel at constant power. Figure 13 uses a constant flux:

Figure 1 -- Reference case showing the basic U-235, time-dependent poison in a hard, constant spectrum (i.e., both flux ratio and thermal spectrum) and at a constant power — The conditions noted on the figure are approximately equivalent to the average values in the PWR Core 2, highly enriched seed (see Figure 19 for a more accurate PWR calculation). As explained, these conditions were taken as a reference basis throughout the course of this investigation from which the importance of changes in or additions to the fission product data were evaluated. (The small superposed perturbation is the result of a 1000-hour zero power interval beginning at $\alpha = 0.78$, or 5000 EFPH.

The return of the resulting transient to the constant power curve is a characteristic property of the fission products. It is a result of the fact that the macroscopic poison of any fission product ultimately attains an equilibrium value such that its depletion rate is equal to its source, and all sources are determined by the power requirement.) The initial increase in RI with depletion probably results from most of the direct yields going to nuclides with small or unknown epithermal cross sections. The later decrease in RI and the over-all decrease in σ_0 results from two dominant factors. First, the main contributors have attained or are approaching saturation so that, as indicated by Equation (11), there is a general tendency for σ_0 and RI to vary as $1/t$. Secondly, because the power is constant and the fuel is depleting, the flux is increasing. Therefore, the destruction rate of the nuclides is slowly increasing while their source is constant. Thus there is a general tendency for the nuclide concentrations to decrease. Both factors are mitigated by the formation of higher order products.

Figure 2. Sensitivity of σ_0 to Flux Ratio B_3 (U-235).

Figure 3. Sensitivity of RI to Flux Ratio B_3 (U-235).

Figures 2 and 3 -- Variations in flux ratio — The conditions are the same as Figure 1 except for the variations in the flux ratio, B_3 . In an actual reactor an increase in flux ratio would normally be accompanied by an increase in thermal hardening (i.e., a decrease in g^4), but in this survey B_3 and g^4 were varied independently. The direction of the changes in σ_0 and RI with an increase in flux ratio is partially explained by the comments under Figures 7 and 8.

Figure 4. Sensitivity of σ_0 to Flux Ratio B_3 without Nd-147 Chains (U-235).

Figure 5. Sensitivity of RI to Flux Ratio B_3 without Nd-147 Chains (U-235).

Figures 4 and 5 -- Variations in flux ratio, no neodymium chain poison -- Curves 4 and 5 are the same as Figures 2 and 3, respectively, except for removal of Nd-147 chain poison. (Refer to Part I, Section IV for a discussion of these chains.) The Nd-147 chains are seen to account for most of the flux ratio dependence of the thermal poison but for only about half of the RI dependence on flux ratio.

Figure 6. Comparison σ_0 and RI with and without ND-147 Chains (U-235).

Figure 6 -- Comparison of Figure 1 with case of no neodymium chain poison -- The Nd-147 chains are seen to account for approximately 30 percent of the thermal poison and 26 percent of the resonance integral (at $\alpha = 0.6$). The importance of these chains depends critically on the flux ratio, flux magnitude, thermal spectrum hardness, and amount of fuel depletion.

Figure 7. Total σ_0 from Nd-147 Chains (U-235).

Figure 8. Total RI from Nd-147 Chains (U-235).

Figures 7 and 8 -- Total σ_0 and RI from the Nd-147 chains as a function of flux ratio; conditions are the same as for Figure 1 -- Since these parametric calculations are all carried out for the same constant power, the thermal flux is decreased and the epithermal flux increased by an increase in flux ratio at any particular depletion. Most of the epithermal poison of this chain (~76 percent) comes from Pm-147* which is predominantly an epithermal absorber; hence, its concentration and, therefore, the total epithermal poison decreases with an increase in flux ratio. The thermal poison is derived primarily from Pm-148m (~27 percent) and Sm-149 (~53 percent), both of which are predominantly thermal absorbers; therefore, a change in the epithermal flux has relatively little effect on their concentrations compared to changes in the thermal flux. Thus an increase in flux ratio, while maintaining constant power, causes an increase in σ_0 and a decrease in RI. In addition to these effects, an increase in epithermal flux significantly increases the competition between β -decay and neutron absorption at the Pm-148 isomer so that a larger fraction of its transmutations reach Sm-149. These comments tend to hold true for the remaining aggregate poison because the nuclides having the largest thermal cross sections generally have relatively small epithermal cross sections, and the converse is also true to a lesser extent. Thus, the aggregate poison would be expected to show the same qualitative spectrum (here, flux ratio) dependences as the neodymium chain, but the dependences are not as marked when the neodymium chain is removed. These points explain most of the directional changes in σ_0 and RI as a result of variations in flux ratio, flux magnitude, and spectrum.

Figure 9. Comparison Sm-149 Poison from Pm-149 and Nd-147 Direct Yields (U-235).

Figure 9 -- Comparison of the Sm-149 production between that produced by the Nd-147 chains and by its production from the Pm-149 direct fission yield -- Late in core life, the Nd-147 chains produce more Sm-149 than is produced by the Pm-149 yield. Potentially, as the flux level is raised so that the (n, γ) reaction rate increases, relative to the β -decay rate, twice as much Sm-149 would be produced by the Nd-147 chains, because the Nd-147 yield is approximately twice as large as the Pm-149 yield. In the PWR-2 reactor the two production modes are nearly equal late in reactor life.

*Refer to Table 3 for the poison distribution of the neodymium chain nuclides in PWR-2.

Figure 10. Sensitivity of σ_0 to Thermal Spectrum, g^4 (U-235).

Figure 11. Sensitivity of RI to Thermal Spectrum, g^4 (U-235).

Figures 10 and 11 — Variations in thermal spectrum factor; conditions are the same as Figure 1 (see note for Figures 2 and 3). Note that, unlike the cases of varying flux ratios, here, both σ_0 and RI decrease when g^4 increases.

Figure 12. Sensitivity of σ_0 and RI to Power Density, P (U-235).

Figure 12 -- Poison for different power densities obtained by increasing the flux level; conditions are the same as for Figure 1 — Thermally or for large α , the cross sections are not strongly dependent on the steady-state power density. This is a fortunate result. As will be seen, however, the magnitude of the poison transient depends strongly on the power density. While the power density can be changed by changing the fuel density, this would not alter the poison at a given α , all other factors being equal.

Figure 13. Sensitivity of σ_0 and RI to (Constant) Flux Level, ϕ^4 ; Flux Ratio, B_3 , is Constant (U-235).

Figure 13 -- Sensitivity of poison to flux level -- In each of the three cases plotted the flux level is maintained at the constant values noted. (The power density is directly proportional to the product $\alpha \phi^4$ and is initially 45.2 w/cm^3 for $\phi = 0.1 \times 10^{14} \text{ n/cm}^2\text{-sec.}$)

Figures 14 through 18 show the transient increase in σ_0 and RI for U-235 fuel after a reduction to zero power following an extended, constant power run:

Figure 14. σ_0 Transients Following Constant Power Runs of 140 w/cm³ (U-235).

Figure 15. RI Transients Following Constant Power Runs of 140 w/cm³ (U-235).

Figures 14 and 15 -- Transients in σ_0 and RI following 140 w/cm³ power runs; conditions are the same as that of Figure 1 -- Also plotted is the transient due to a zero power interval of 1,000 hours duration beginning at 5,000 EFPH. This transient has also been superposed on the constant power plots versus α of Figure 1.

Figure 16. σ_0 Transients Minus Nd-147 Chains (U-235).

Figure 17. RI Transients Minus Nd-147 Chains (U-235).

Figures 16 and 17 -- Transients without the neodymium chain; 140 w/cm³ -- These plots are otherwise identical to Figures 14 and 15, respectively. The magnitude of the epithermal transient has increased considerably, but the thermal transient has been reduced more than 50% in magnitude.

Figure 18. σ_0 and RI Transients Following Constant Power Runs of 560 w/cm³ (U-235).

Figure 18 -- Transients in σ_0 and RI following 560 w/cm³ power runs to compare with Figures 14 and 15 -- Conditions are the same as Figure 1, but the power density has been arbitrarily, and unrealistically, increased by a factor of four. In this case the massive transient in the thermal poison (~50% increase over the value at constant power) is primarily the result of the decay of (53.1h) Pm-149 to Sm-149 in the Nd-147 chains. The epithermal transient is not so greatly increased. A calculated increase in the transient poison is expected at this higher power density because the steady-state concentration of the radioactive nuclides is larger; these contribute comparatively little to the poison at steady-state power while they decay to nuclides with relatively large cross sections when the power is reduced. The stable nuclide concentrations are relatively insensitive to an increase in power (i.e., flux level) since both their source and depletion rate increase. An increase in power density does favor the coupling by neutron absorption over that of radioactive decay in the radioactive nuclides. The effect of this on σ_0 and RI is not readily apparent since a change in the coupling will increase the poison in some chains and decrease it in others. However, it would be expected that this effect would be small (as indicated by the power variations of Figure 12).

Figures 19 through 23 contrast the fission product poison in the PWR-2 highly enriched seed and a typical natural uranium blanket cluster. While the seed values are plotted versus fuel fraction and the blanket values versus MWD/TONNE, the plots shown cover the same degree of reactor lifetime. The actual variation in flux over reactor lifetime, as determined by spatial calculations, was used; beginning and end-of-life values are noted:

Figure 19. Average PWR-2 Seed σ_0 and RI (Highly Enriched).

Figure 19 -- PWR-2 seed poison — The average seed fluxes and fuel density was obtained from a TURBO (Reference 7) depletion calculation.

Figure 20. Typical PWR-2 Blanket σ_0 and RI (Natural UO₂).

Figure 20 -- PWR-2 blanket poison — This represents a typical PWR-2 blanket cluster near the highly enriched seed. σ_0 and RI are about 13 percent larger thermally and 18 percent epithermally here than for corresponding seed values late in reactor life. The reason is partly due to the greater yield fractions of several of the large fission product poisons from the Pu-239 and U-238 fuels than from U-235, and partly to spectrum differences. The different yield fractions are believed to be the main factor as indicated in Figure 21.

Figure 21. Comparison of σ_0 and RI from U-235 and Pu-239 (PWR Blanket Spectrum).

Figure 21 -- Comparison of U-235 and Pu-239 poisons — These calculations used the same spectrum and over-all flux history of the blanket calculation of Figure 20 to irradiate identical initial concentrations of U-235 and Pu-239. After 16×10^{19} fissions/cm³ (≈ 5000 hours irradiation in the blanket flux) the Pu-239 poison exceeds the U-235 value by 34 percent in RI and 31 percent in σ_0 . Most (~ 85 percent) of the poison difference can be attributed to nine nuclides. These and the differences in their poisons in percent of the total U-235 poison are shown in Table 2. No unlisted nuclide represents more than 1.8 percent of the remaining difference. As shown in Figure 21, the percentages depend on the number of fissions.

TABLE 2. DIFFERENCE IN NUCLIDE POISONS PRODUCED BY Pu-239 AND U-235 IN PERCENT OF TOTAL U-235 FISSION PRODUCT POISON

Nuclide	% σ_0	% RI
Sm-151	9.9	--
Rh-105	9.5	--
Rh-103	4.8	8.4
Nd-143	-6.2	--
Eu-155	2.6	--
Sm-152	2.4	6.6
Ag-109	2.2	9.3
Eu-153	1.9	--
Xe-131	<u>1.7</u>	<u>2.9</u>
	28.8	27.2
Total % Difference	31	34

Figure 22. σ_0 and RI in PWR-2 Seed without Nd-147 Chains.

Figure 23. σ_0 and RI in Typical PWR-2 Blanket without Nd-147 Chains.

Figures 22 and 23 -- PWR-2 seed and blanket, respectively, without the Nd-147 chains. Otherwise, these are identical to Figures 19 and 20.

Figures 24 and 25 are included for general interest:

Figure 24. σ_0 and RI for Natural UO_2 in Soft Spectra.

Figure 24 -- Natural UO_2 in a soft spectrum characteristic of the MTR. A constant flux was used.

Figure 25. σ_0 and RI for Slightly Enriched ($N^{25}/N^u = 3.4\%$) Uranium.

Figure 25 -- Slightly enriched (3.4 percent) uranium reactor, hard spectrum. Fluxes were taken from a CANDLE (Reference 7) depletion calculation.

In view of the above results and comments, the qualitative dependence of σ_0 and RI on fuel depletion, flux ratios, etc., is readily understandable. Historically it was assumed that the aggregate fission product cross section per fission would be nearly constant and independent of the particular reactor environment in which it is generated. Such is clearly not the case. While the variations in the cases considered are not particularly large, many are nonetheless too large to ignore, particularly for a long-lived reactor or for a reactor which has a substantial fraction of its reactivity control predetermined by its nuclear design so that it is not subject to control during operation (e.g., by a burnable poison). The greatest variation in σ_0 and RI is that accompanying fuel depletion. It is certainly too large to be ignored. Whether or not the other variations are important will depend on particular reactor design objectives.

It has already been noted that most of the thermal transient characteristics and a large fraction of the total poison is due to the neodymium chain discussed in Part I, Section IV (primarily, linear chains 56 and 57 of Table 1). In Table 5, the 14 most important of the 67 chains (in magnitude and transient response) are listed including chains 56 and 57. These also account for all significant transient behavior of the gross poison. Of those listed, chains 22, 45, 56, and 57 (epithermally only) show a significant decrease in poison following a power reduction. The poison from chains 22 and 45 at $\alpha = 0.96$ on Figure 14 (1000 EFPH of PWR-2) decreases thermally more than the Sm-149 increases in the neodymium chains. This is because of Rh-105 and Xe-133, and if their cross sections are much in error the early-in-life transient behavior will (thermally) be much more significant than shown in Figure 14. By $\alpha = 0.78$ (5000 EFPH) the significant transient behavior is due to the neodymium chain, and interest in transients can be confined to it.

The early-in-life PWR transients of natural uranium are very similar in magnitude and direction to those of the U-235 seed. Later (>3,000 hours) the thermal transient is about 3 percent less and the epithermal about 1 percent more than the corresponding seed values. The main reason for the smaller thermal transient is (36h) Rh-105 in chain 22, which has a large Pu-239 yield. Its decrease in thermal poison slightly exceeds the increase from the neodymium chain. The epithermal transients are not very significant in either case.

Because most of the poison dependence on the flux magnitude, ratio and spectrum, and the transient response to power variations is due to the neodymium chain, they have been singled out for emphasis in many of the calculations reported. However, the reader should be aware that the large magnitude of the neodymium chain poisons is contingent on the accuracy of the Pm-148 cross sections (Reference 11), for these are in competition with β -decay. As noted for Figure 6, these chains may account for 25 to 30 percent of the aggregate poison. The (5.39d) Pm-148 isomer and its progeny account for ~4 percent of the aggregate poison. The cross sections of this isomer have been determined only to within about ± 60 percent. This uncertainty in cross section generates a nearly equal uncertainty in the Pm-148 poison or ~2.4 percent of the aggregate poison, because its saturation level is determined strongly by its half-life. The (40.6d) Pm-148m is, of course, the more important isomer since it and its progeny generate over 80 percent of the neodymium chain thermal poison. Its cross section is quoted with an uncertainty of ± 18 percent. However, this will create only about 3.5 percent error in its poison or ~1 percent in the aggregate poison, because its macroscopic poison is determined mostly by its neutron absorption rate, rather than the radioactive decay rate. (It will be seen in Part II, Section V that when a fission product density is determined only by its neutron absorption and source rate that its effective macroscopic poison at saturation is independent of its cross section. These statements on uncertainty apply only at saturation.) These values, of course, are approximate since they depend on the flux level and on the irradiation time.

In addition to these uncertainties, the cross sections of Nd-147 and Pm-149 are unknown. If their cross sections are larger than approximately 3000 barns, then the branching at these nuclides would be significantly altered. It was pointed out in Reference (11) that the calculated value of Pm-149 production was approximately twice the value determined by activation measurements. The quoted uncertainty in the Pm-148 cross sections does not account for this discrepancy. However, an effective Pm-149 cross section of about 7000 barns would explain the difference. Such a cross section would not change the total poison generated by the neodymium chains, but it would greatly reduce the calculated transients at high power densities (e.g., that of Figure 18). An additional confirmation of the Pm-148 cross sections and a measurement of the Nd-147 and Pm-149 cross sections is clearly needed.

The percent of the total neodymium chain poison from each nuclide is shown in Table 3 for the PWR-2 seed at 10,000 EFPH ($\alpha = 0.562$).

The values apply to the seed, but the corresponding blanket values are not significantly different.

TABLE 3. PERCENT OF TOTAL NEODYMIUM CHAIN POISON FROM EACH NUCLIDE
(SEED 10,000 HOURS)

Nuclide	% Thermal (σ_o)	% Epithermal (RI)
Pm-147	14.2	71.6
Pm-148m	25.5	10.9
Pm-148	1.0	11.3
Sm-149	49.4	0.6
Sm-150	2.7	5.2
Sm-151	7.1	0.4

Total chain RI = 45.6 barns/fission = 24% of total FP poison.

Total chain σ_o = 16.4 barns/fission = 26% of total FP poison.

II. ORDERED LIST OF IMPORTANT NUCLIDE POISONS AND REDUCED REPRESENTATIONS

In Table 4 the most important individual nuclide poisons generated by U-235 fission are identified. The table is based on the conditions of Figure 1 ($B_3 = 2.5$, $g^4 = 0.5039$, $P = 139.8 \text{ w/cm}^3$), and the nuclides are listed in descending order of their thermal poison at 5000 EFPH ($\alpha = 0.78$). Enough nuclides are listed in this manner to include ~95 percent of the total poison in both energy groups at three times in reactor life (1000, 5000, and 10,000 EFPH). The columns list the percent poison due to each nuclide at the indicated lifetimes. The next largest of the nuclide poisons not listed is noted for each column. Thus, in no case is there any unlisted poison which exceeds 1.4 percent of the total group poison. The relative importance of these nuclides depends on the degree of fuel depletion, as indicated by comparing the columns at the three values of α given in the table.

The ordering also depends on the type of fuel. In the case of natural uranium, Ag-109 and Eu-155 assume a much greater importance than is indicated in Table 4. Also, if Pd-107 possesses a thermal cross section of about 375 barns (only an upper limit of ~750 b is known), then it would add an additional

TABLE 4. PERCENT TOTAL POISON FROM INDIVIDUAL NUCLIDES*

Nuclide	$\alpha = 0.9558$		$\alpha = 0.7800$		$\alpha = 0.5619$	
	← 1000 →		← 5000 →		← 10,000 →	
	EFPH		EFPH		EFPH	
	Gp. 3	Gp. 4	Gp. 3	Gp. 4	Gp. 3	Gp. 4
Nd-143	2.56	10.29	3.45	19.96	3.70	22.56
Sm-151	3.8	42.77	1.14	18.39	0.72	12.28
Sm-149**	0.10	5.55	0.23	17.92	0.20	16.74
Pm-148m	2.66	4.07	3.87	8.53	3.16	7.35
Pm-147	25.52	3.29	23.41	4.34	17.49	3.42
Xe-131	9.59	2.61	9.99	3.92	10.10	4.18
Rh-103	4.79	1.26	9.61	3.64	10.85	4.33
Nd-145	5.26	2.34	4.41	2.82	4.70	3.17
Cs-133	12.30	1.54	12.38	2.24	13.23	2.52
Rh-105	1.43	6.54	0.24	1.60	0.13	0.91
To-99	6.62	1.18	6.05	1.56	6.55	1.70
Sm-152	5.29	0.87	6.06	1.44	6.06	1.52
Eu-155	0.64	2.92	0.21	1.41	0.29	2.03
Eu-150	1.23	0.74	1.59	1.36	2.42	2.17
Kr-83	0.64	1.09	0.53	1.30	0.56	1.44
Sm-150	2.14	0.72	2.67	1.29	3.52	1.36
Pd-107†	0.15	0.69	0.12	0.81	0.12	0.86
La-139	0.39	0.57	0.33	0.71	0.37	0.83
Pr-141	0.26	0.23	0.51	0.65	0.64	0.87
Eu-154	0.02	0.08	0.10	0.63	0.27	1.88
Pr-143	2.55	2.17	0.51	0.62	0.28	0.36
Xe-133	0.51	2.33	0.09	0.58	0.03	0.34
Mo-95	0.17	0.04	1.26	0.44	2.22	0.81
Nd-146	0.19	0.30	0.16	0.30	0.10	0.48
Nd-144	0.31	0.28	0.28	0.36	0.33	0.45
Ru-101	2.08	0.25	1.78	0.30	1.95	0.35
Pm-148	2.41	0.15	3.12	0.28	2.97	0.28
Cs-135	0.72	0.19	0.59	0.22	0.60	0.23
Mo-97	0.47	0.13	0.40	0.16	0.44	0.19
Kr-82	0.30	0.12	0.25	0.15	0.27	0.17
Sm-147	0.07	0.02	0.39	0.12	0.65	0.22
Pd-105	0.31	0.08	0.28	0.10	0.30	0.12
Zr-93	0.98	0.07	0.84	0.09	0.93	0.10
Zr-91	0.08	0.03	0.16	0.07	0.22	0.11
Ru-102	0.23	0.06	0.20	0.07	0.22	0.09
Nd-148	0.45	0.06	0.38	0.07	0.42	0.08
Mo-100	0.22	0.03	0.19	0.04	0.21	0.05
Ag-109	0.23	0.03	0.18	0.03	0.19	0.03

TABLE 4 (Cont)

Nuclide	$\alpha = 0.9558$		$\alpha = 0.7800$		$\alpha = 0.5619$	
	1000		5000		10,000	
	EFPH		EFPH		EFPH	
	Gp. 3	Gp. 4	Gp. 3	Gp. 4	Gp. 3	Gp. 4
Xe-132	0.20	0.01	0.19	0.01	0.23	0.02
Cs-137	0.39	0.01	0.33	0.01	0.37	0.01
%	98.26	95.68	98.46	98.63	98.11	96.65
Largest	Mo-98	Cd-113	Mo-98	Cd-113	Mo-98	Cs-134
% not listed	0.19	1.40	0.16	0.37	0.18	1.05
	RI	σ_o	RI	σ_o	RI	σ_o
Total b/f	184.45	101.61	214.95	82.21	193.72	70.30

*Based on constant power calculation: $P = 139.82 \text{ w/cm}^3$

$$\phi^3/\phi^4 = 2.5$$

$$N_o^{25} = 4.27 \times 10^{20} \text{ at/cm}^3$$

**Does not include production from Pm-149 yield.

† $\sigma_{2200} = 375\text{b}$ and $\sigma^{\text{epi}} = 16.4\text{b}$ was assumed for Pd-107 in these calculations and those of Table 7, but the values listed in Table 1 were used in all other calculations reported herein.

6 percent to the thermal poison. Its progeny, Pd-108 must then be added to the table. Such a Pd-107 cross section would add no more than 1 percent to the U-235 poison. The remaining nuclides listed do not significantly change in ordering for natural uranium.

The nuclides and chains listed in Table 1 do not all produce a significant poison. They are intended to be a complete description of the fission products as they are presently known. From a more practical standpoint, many of the 67 chains could be omitted and many others produce a nearly constant number of barns/fission. Those chains producing a constant barns/fission can be represented by a single pseudo-nuclide. With the aid of the important nuclides listed in Table 4, 14 of the 67 chains containing 104 of the 256 nuclides listed in Table 1 have been identified which produce approximately 95 percent of the total poison. The percent each chain contributes to the total poison is given for three fuel fractions in Table 5 for U-235 fuel. The remaining ~5 percent of the poison is relatively constant and can be represented by the following pseudo-nuclide parameters:

σ_{2200}	σ^{epi}	% Yields		
		U-235	U-238	Pu-239
4.10	1.21	107.9	100.8	92.90

The yields are the actual totals for the remaining 53 chains which this pseudo-nuclide represents. The cross sections are small enough so that this pseudo-nuclide will not saturate for flux-times less than about 10^{23} n/cm^2 .

TABLE 5. FOURTEEN CHAIN REPRESENTATION, PERCENT OF
TOTAL POISON PER CHAIN*

Chain No. (Table 1)	$\alpha = 0.9558$ 1000 EFPH		$\alpha = 0.7800$ 5000 EFPH		$\alpha = 0.5619$ 10,000 EFPH	
	Group 3	Group 4	Group 3	Group 4	Group 3	Group 4
17	0.89	0.21	1.88	0.64	2.90	1.05
19	13.91	2.84	17.75	5.72	19.70	6.86
22	1.76	6.85	0.52	1.73	0.43	1.04
24	0.38	0.09	0.31	0.10	0.31	0.11
43	9.82	3.13	10.07	4.09	10.15	4.30
45	0.64	2.52	0.19	0.73	0.16	0.51
46	12.47	1.64	12.51	2.67	13.42	3.62
55	10.52	15.33	8.42	23.78	8.72	26.54
56	28.88	10.53	28.35	26.65	22.40	24.97
57	2.49	1.21	3.39	4.72	3.52	5.52
63	2.08	1.13	3.06	1.28	4.18	5.97
64	10.45	45.26	8.39	18.75	8.17	13.32
65	0.65	3.06	0.16	1.02	0.07	0.45
67	0.02	1.39	0.01	0.28	~0	0.12
Total %	94.96	95.19	95.01	95.16	94.14	94.38

*Based on constant power: $P = 139.82 \text{ w/cm}^3$

$$\phi^3/\phi^4 = 2.5$$

$$N_0^{25} = 4.27 \times 10^{20} \text{ at/cm}^3$$

The 14 chains plus the pseudo-nuclide will duplicate the results of the 67 chains in Table 1 within ± 1 percent after approximately the first 100 hours of full power operation. However, for spectra and flux-times which differ considerably from any of the values used in this report all of the chains should be used.

The 14 chains were actually selected because their poison changes during irradiation, but they also produce the bulk of the poison. Chain 24 can clearly be exempted; it was selected on the basis of earlier calculations which used a large Pd-107 cross section.

No attempt has been made toward an even simpler representation, but depending on the maximum flux-time and the accuracy sought, simpler representations are clearly possible. For example, in chain 43 (one of the 14 important chains) the yield fractions can be moved to Xe-131, and Xe-132 can be omitted from the end of the chain with no significant loss in poison for most power reactors. Thus a four nuclide chain is reduced to a single nuclide description. For U-235 fuel, chain 67 can be eliminated altogether with a 2 percent maximum, early-in-life loss in poison. Others can be grouped into the pseudo-nuclide above to good approximation. Similar, and equally obvious, manipulations can be made on most of the

remaining chains, but the shortening of chains in the manner described for chain 43 will depend largely on the reactor to which they will be applied and on the desired accuracy. (In this regard, the reader is reminded of the discussion in Part I, Section III. The chain termination will depend on the maximum flux-time anticipated. For instance, the elimination of Sm-151 from chain 56 would alone create an error of about 2 percent in aggregate poison late in the PWR-2 lifetime. Also, its progeny up to Eu-155 all contribute a significant poison. Since these are 7th or higher order fission products, this clearly indicates that neutron capture in second and higher order fission products cannot be ignored as some investigators are prone to do. Thus, even if all products were stable as generally assumed, some (n, γ) chains would require an 11th to 15th order deflection scheme.)

Through careful consideration of the nuclides which produce the bulk of the poison, particularly with respect to end use of the poison, the number of nuclides which must be explicitly described can undoubtedly be reduced considerably. For limited use it appears that it would be practical to incorporate an explicit, pointwise spatial calculation of the fission products into a one-dimensional depletion program such as CANDLE (Reference 7). For instance, the actual "one-point" CINDER calculation time, exclusive of read-in and read-out time, is 0.00009 min/nuclide/timestep for the 67 chains of Table 1 and 0.00014 for the 14 chains. (The calculation time per nuclide is a function of the length of the chains and is therefore longer for the 14 than for the 67 chains, since the latter contains many short chains.) Even for the 104 nuclides in the 14 chains mentioned, a 100-point depletion calculation would require only 1.5 minutes computation time per timestep. This is still a long computation time, but the 14 chains are also a very detailed representation of the fission products. Very likely the number of nuclides could be reduced to 50 or less, and the length of chains shortened considerably. Also, by selecting those chains among the 14 which have similar characteristics and fitting each group by a single fictitious chain, the calculation time can be reduced to insignificance.

It is not yet apparent that an explicit pointwise representation is necessary. At the present time the PWR design procedure uses average region fluxes in the CINDER program to obtain aggregate cross sections which are then used in each fuel region and at each timestep in depletion calculations. This procedure is believed to be quite adequate for reactivity calculations.

III. OMISSION OF RADIOACTIVE DECAY FROM THE AGGREGATE FISSION PRODUCT POISON CALCULATIONS

Most treatments of fission product poisoning have ignored the detailed effects of radioactive decay by treating only the stable or near stable nuclides, that is, by assigning the isobaric yield fractions to such nuclides directly. There is some interest in knowing the error associated with this approximate approach. It is obvious that β -decay must be explicitly used in describing the Nd-147 chains. Therefore, any stable nuclide treatment must exclude these chains; they must be treated separately as a special case. (It should be noted that the effect of radioactive decay cannot be evaluated by the simple expedient of setting all λ 's equal to zero, for this would prevent the formation of most important stable nuclides.)

The treatment used by Greenhow and Hansen (References 3 and 4) appears to be the best approach short of including all coupling systematics and will be used for comparison. They have treated all nuclides with half-lives greater than approximately 50 hours as stable and those with half-lives less than 50 hours as instantaneously decaying. The placement of the yield fractions was correspondingly adjusted. The "stable" nuclides are thus coupled only by neutron absorption. (The retention of (n, γ) couplings is very necessary as indicated by the poison associated with 7th and higher order products in the last section.)

This approach automatically eliminates the branching problems, and time is no longer an independent variable in the nuclide concentration. However, there are four relatively distinct sources of error:

(1) nuclides with half-lives less than 50 hours cannot contribute to the poison, (2) only the nuclide poisons due to neutron capture can be (approximately) followed, (3) the poison from all radioactive nuclides which are assumed stable is overestimated (for λ , if present, would decrease the calculated nuclide density, and (4) there can be no transients. The second error underestimates and the third error overestimates the aggregate poison so that it may not be grossly in error after a large fuel depletion, although the individual nuclide poisons may be in considerable error.

In order to evaluate these errors collectively, a comparison of the present results with that obtained by Greenhow and Hansen is suggested. However, the cross sections used by them are different in many cases from the later compilation used in this report. Also, several nuclides used here do not appear in their compilation. Therefore, the effect of radioactive decay alone cannot be determined by a simple comparison of calculations. Instead, it appears that a simple, meaningful comparison can be made by using the cross sections and chains of "stable" nuclides reported earlier by Greenhow and Hansen (Reference 4) in one case, and then, using only the same nuclides and cross sections to reform the chains as demanded by radioactive decay and to perform the calculations again for the same irradiation history.

This comparison was made for the reactor conditions noted on Figure 1. It was found that the error in the thermal aggregate cross section varies from +32 percent at 5-percent depletion to +7 percent at 40-percent depletion. The error in the epithermal value varies from +39 percent at 5-percent depletion to +19 percent at 40-percent depletion.* The Xe-135, Sm-149, and Nd-147 chain poisons previously discussed were not included in either calculation. This relative comparison clearly shows that radioactive decay cannot be readily excluded, if at all.

The tendency of the two calculations to approach each other at high depletion is expected because of the saturation of the radioactive nuclides. However, there could not be a convergence even apart from the radioactive branching because the presence of radioactive decay in a nuclide reduces its concentration even at saturation, i.e., at saturation

$$\frac{\lambda}{\sigma} = \frac{Y\bar{\sigma}}{(\lambda + \bar{\sigma}\phi)t},$$

whereas the stable treatment omits the λ . There is, however, no reason why the decay constant must be excluded, even when only (n, γ) coupling is permitted.

IV. COMPARISON WITH EXPERIMENT

An experimental investigation of U-235 fission product poisoning is being carried out by S. B. Gunst, et al (Reference 13) as a part of the PWR Physics program in the experimental determination of long-term reactivity changes in irradiated fuels. At the present time samples of enriched uranium in zirconium have been exposed to a fuel depletion of ~87 percent in the Materials Testing Reactor (MTR). Periodically, during the irradiation the samples are removed for measurement in the Reactivity Measurement Facility (RMF) or the Advanced Reactivity Measurement Facility (ARMF). Using few-group perturbation theory,

*This is an evaluation of the inherent validity of the stable nuclide type calculations. The extensive chains and higher order poisons and, in particular, the neodymium chain used in the present results add enough to the poison at large depletions to reverse the thermal comparison, making the thermal values reported here 20 to 30 percent larger than the values reported in Reference (4).

reactivity changes can be associated with group cross sections. Changes in reactivity due to the heavy (fuel) isotope constituents and to the Xe-135 and Sm-149 poisons (from the direct fission yields to the mass 135 and 149 chains) are calculated from a knowledge of the pre-irradiation constituents of the sample and flux histories inferred from cadmium-covered and bare Co-59 monitors. The remaining reactivity changes are due to the gross fission products and are interpreted in terms of a thermal and an epithermal cross section. The experimental details and some preliminary results for Zircaloy-2 clad samples of highly enriched uranium in zirconium are available in Reference (13). Uncertainty associated with measurement statistics is ± 10 percent, but a more accurate assessment of uncertainties has not been assigned to date because of the possibility of systematic errors still under investigation.

A comparison of the experimental results with the analytical treatment described in this report has been made for a sample for which the experimental results were considered to be particularly accurate. Neutron fluxes were inferred from cadmium-covered and bare Co-59 monitor activations; the monitors, replaced after each three-week cycle of MTR irradiation, provided measures of the epithermal and thermal integrated flux-times at three-week intervals. A small correction was applied to the activations to simulate a Cd-cutoff at 0.625 ev. A larger correction, accounting for the variation in thermal flux level between the position of the monitors and that of the sample, was determined experimentally and supplied with the monitor data. Fluxes were constructed on the assumption that a KATE description (Reference 12) of the MTR spectrum applied below 0.625 ev and that the spectrum was $1/E$ above this energy.

The results of the calculation are shown in Figures 26 and 27. The average fluxes, inferred for each three-week interval for which the sample was irradiated, are plotted as a histogram in Figure 28. The variations in the flux level are partly the result of the sample removal from the MTR for measurement in the RMF before completion of a cycle. The thermal transients evident in Figure 26 are due to the zero flux intervals which extended over an entire cycle or more. The epithermal transients are minor.

The experimental determination of barns/fission has, to date, been made at 64 and 81 percent depletion. The experimental numbers obtained are based on a division between the thermal and epithermal energy groups at $E = 0.12$ ev. Therefore, a correction must be applied to the calculated RI which cutoff at 0.625 ev to account for the cross section above 0.12 ev. This was done by assuming that the aggregate cross section below 0.625 ev was $\sqrt{0.0253} \sigma_0 E^{-1/2}$ and that the spectrum between the two cutoffs was $1/E$. The correction is

$$\Delta RI = \sigma_0 \sqrt{0.0253} \int_{0.12}^{0.625} dE/E^{3/2} = 0.516 \sigma_0 . \quad (19)$$

This correction adds only ~ 10 percent to the calculated RI and cannot introduce a large uncertainty.

The experimental values represent an average of the determinations following the removal of the sample from the MTR; most measurements were made near the maximum of the transients evident in Figure 26. Averaging the transient values occurring at the approximate measurement times and correcting the RI as indicated above, the corresponding experimental and calculated values are as shown in Table 6.

TABLE 6. COMPARISON: EXPERIMENTAL AND CALCULATED
FISSION PRODUCT CROSS SECTIONS
(RI From 0.12 ev)

Exp.	Calc.	Depletion
σ_o : 55	54	64%
RI: 210	236	
σ_o : 47	18	81%
RI: 190	220

Figure 26. Thermal Cross Section (σ_o)
for Experimental Sample.

Figure 27. Resonance Integral (RI) for
Experimental Sample.

Figure 28. Flux Histogram for Experimental
Sample.

Since the present calculations indicate that the fission product poison depends significantly on the reactor environment in which it is generated, a measurement of the poison generated in a particular environment is of little use to the nuclear designer; instead, he must rely on the calculational model. Therefore, considerable effort should be expended to find the cause of any discrepancy between calculation and experiment so that the nuclear designer can use calculations with confidence. At the present time the agreement exceeds all expectations. However, the reader should not take this remarkable agreement to be a confirmation of the adequacy of the presently known physical data. No complete

assessment of experimental uncertainties has yet been made. Also, similar comparisons in other spectra, at other power densities, and at other depletions are desirable. The thermal poison generated in a harder spectrum is expected to be significantly larger (compare Figures 2 and 10). A hard spectrum experiment is now in progress, but results are not yet available for comparison.*

V. DISCUSSION OF ERRORS

Because the calculational method outlined in Part I includes all coupling mechanisms and, potentially, all spectrum effects, it does not introduce errors. The probable error depends primarily on the adequacy of the available physical data.

Several factors influence the uncertainty in the calculations reported:

1. Uncertainty in the cross section
2. Uncertainty in the yield fractions
3. Assignment of the direct yields to particular nuclides in the mass chains
4. Uncertainty in half-lives of the radioactive nuclides
5. Irradiation time
6. Flux magnitudes and spectra
7. Use of a $1/E$ flux in averaging the resonance cross sections
8. Assumption that the thermal cross section is $1/v$
9. Self- and mutual-shielding

Items 5 and 6 make an error study of general significance virtually impossible. Apart from the obvious difficulty in assessing the total uncertainty resulting from the above factors, a detailed assessment is somewhat meaningless and misleading because there is a possibility that the as-yet-unmeasured cross sections may significantly alter the aggregate poison, and this factor cannot be completely evaluated. However, an approximate evaluation of the above factors, particularly as it indicates their relative importance, seems to be warranted. Some of these factors are not nearly as important as it might be expected from a cursory examination.

The nuclides listed in Table 4 contribute ~95 percent of the total calculated poison. For these, most of the individual 2200 m/s cross sections are known to $\sim \pm 5$ percent. The uncertainty in the epithermal cross sections varies widely, but the average, weighted by the percent contribution to the total poison listed in Table 4, is less than ± 20 percent.** The probable error of the total poison as a result of these uncertainties is, of course, much less. Among other things its value depends on the magnitude of the epithermal-to-thermal flux ratio and the thermal spectrum hardness. Thus the expected error in the effective aggregate poison increases with increasing flux ratio, because this enhances the importance of the resonance cross sections. However, for any flux spectrum it should be

*Lack of, or incorrect, cross sections affect the accuracy of calculated transient changes in poison more than that of the poison at power. This hampers meaningful comparisons with the experimental values of samples irradiated at high power and measured at zero power. A recent comparison for a sample irradiated in a flux approximately four times that in Figure 28 showed considerable disagreement, apparently due entirely to the normal Sm-149 transient and its transient in the Nd-147 chain. The results, however, are preliminary. The Pm-149 cross section, suggested in Part II, Section I, would considerably reduce the calculated transient at high power and eliminate the disagreement while neither changing the steady-state poisons in this report nor reducing the values in Table 6 by more than 1 percent.

**The uncertainty associated with the neodymium chains was discussed in Part II, Section I.

kept in mind that the cross sectional error is most important in the determination of the aggregate poison only at the beginning of reactor life where there is no significant poison. After a few thousand hours of irradiation any errors in the direct fission yields appear to dominate the total error. In order to see this, consider the limit of Equation (9). At $t = 0$ the limit, in terms of the effective cross section (Equation (7)), is:

$$\frac{\Delta}{\bar{\sigma}}(0) = y \bar{\sigma} , \quad (21)$$

and the errors in y and $\bar{\sigma}$ are equally weighted. However, at saturation

$$\frac{\Delta}{\bar{\sigma}} = \frac{y \bar{\sigma}}{(\lambda + \bar{\sigma} \phi) t} . \quad (22)$$

Most of the important fission products have small or zero λ 's compared to $\bar{\sigma} \phi$ (Pm-148 excepted). Then,

$$\frac{\Delta}{\bar{\sigma}} = \frac{y}{\phi t} , \quad (23)$$

and the cross section has no effect on $\frac{\Delta}{\bar{\sigma}}$; it serves only to determine the saturation time. Also, it should be kept in mind that most of the important fission products have large cross sections and, therefore, saturate early in reactor life. Thus, one can make the seemingly paradoxical statement that the larger the nuclide cross section, the lesser its importance in assessing the uncertainty in its poison. Higher order nuclides and nuclides with moderate cross sections but with large yields probably introduce most of the poison uncertainty in very long lived reactors.

The maximum probable error in the aggregate poison resulting from cross sections of the nuclides listed in Table 4 was computed for PWR-2 at mid-life (5000 EFPH):

TABLE 7. MAXIMUM PROBABLE ERROR IN POISON
DUE TO CROSS SECTIONS

<u>% Seed</u>	<u>% Blanket</u>
σ_0 2.3	7.8*
RI 7.1	6.3

*For these calculations it was assumed that the Pd-107 thermal cross section is 375 ± 375 b, and this accounts for most of the probable error in blanket poison. All other results used the 10b value as noted in Table 1.

The microscopic cross-sectional errors used were mainly those reported by Garrison and Roos (Reference 5). In this computation it was assumed that each group cross sectional error was reflected as a proportionate error in the nuclide poison. Therefore, the values are a maximum, never obtained, but they are exclusive of any significant unmeasured nuclide cross sections.

This discussion ignores the error propagated from parent to daughter of the important nuclides. Very often the daughters have small cross sections and will attain importance only after a long irradiation time. Therefore, it appears that the error in the aggregate poison is relatively large at small

irradiation times (refer to the discussion in Part I, Section III) and also after very long irradiation times, whereas the minimum error occurs at some intermediate time.

The yield fraction for the important nuclides have been measured mostly by the mass-spectrograph and are accurate to $\sim \pm 3$ percent; those yields determined by radio-chemical data are accurate to ± 10 percent (Reference 10). From the above discussion it is apparent that while the microscopic cross sectional uncertainties are generally larger than uncertainties in yields, their effect on the uncertainty in the aggregate poison is probably comparable.

The division of the effective poison between the thermal and epithermal groups is less certain. The saturation argument above applies only to the effective poison, Equation (7). Even for such saturated nuclides, the division of the poison between the thermal and epithermal groups depends on the relative magnitude of the individual thermal and epithermal cross sections. Fortunately, the poison defined by Equation (7) most nearly determines the effect of the fission products on reactivity.

Unless there was evidence to the contrary in Katcoff's assignment of the cumulative yields (Reference 10), the yield fractions used in this report were placed on the first precursor retained in the linear chains. The shortest lived nuclides used in this analysis have half-lives ~ 10 hours; for such half-lives this assignment of the yields is supported by studies of nuclear-charge distribution in fission (Reference 14).

Half-lives are generally quoted to two and three significant figures, and they are, therefore, accurate between 0.05 and 5 percent. The effect of this on the aggregate poison is expected to be negligible compared to that of the yield and cross sectional error.

In general, the large contributions to the thermal poison are not $1/v$ cross sections as has been assumed in this report. However, by the analysis above, these saturate early in reactor life after which the effective poison is independent of the nuclide's cross sections. For example, consider Sm-149 which possesses a highly non- $1/v$ cross section (40,800 barns at 2200 m/s). When this was increased by ~ 300 percent its thermal macroscopic poison changed only 3 percent after $\sim 10,000$ hours of irradiation (conditions are the same as in Figure 1). The effective poison increased only 0.09 percent. The time of saturation is altered. Thus at 1000 hours the effective poison in the two cases differ by ~ 12 percent. It is clear from this extreme example that the $1/v$ treatment of all of the cross sections will probably introduce negligible error in the aggregate poison. (The results reported here used the correct hardened Sm-149 cross sections.) An examination of the non- $1/v$ contributors indicates that the error is no more than ± 2 or 3 percent. The correct concentration of a nuclide and transients following power changes do depend on the cross section. For this reason and in order to improve the accuracy of calculations at all irradiation times, it is desirable to use the correct spectrum-averaged cross sections. The remaining non- $1/v$ cross sections of importance are now being incorporated into the KATE code (Reference 12), to facilitate the calculation of hardened values for use in CINDER.

A cursory examination of self- and mutual-shielding due to the buildup of nuclides having a large resonance has also been made. The possibility of any significant shielding was found to be unlikely for the flux-times used in this report. Also, as with the non- $1/v$ cross sections, any cross sectional errors due to shielding tend to drop out of the fission product poison calculation.

The use of a $1/E$ spectrum in averaging the epithermal cross sections has not been evaluated. In many cases only a resonance integral was known so that an accurate assessment is not possible. The associated uncertainty, however, is expected to be small for water moderated reactors.

Of the 163 nuclides included in this analysis there are 31 for which no cross sections were available: (49h) Zn-72, (14.1h) Ga-72, (38.7h) As-77, (65d) Zr-95, (35d) Nb-95, (66h) Mo-99, (39.7d) Ru-103, (7.6d) Ag-111, (43d) Cd-115m, (53h) Cd-115, (27.5h) Sn-121, (2.8d) Sb-122, (136d) Sn-123, (60d) Sb-124, (9.6d) Sn-125, (91h) Sb-127, (105d) Te-127m, (37d) Te-129m, (30h) Te-131m, (77h) Te-132, (20.8h) I-133, (6.7h) I-135, (13d) Cs-136, (33d) Ce-141, (11.1d) Nd-147, (53.1h) Pm-149, (47h) Sm-153, (28.4h) Pm-151, (15.4d) Eu-156, (15.4h) Eu-157, and (18h) Gd-159. The uncertainty associated with these nuclides was briefly discussed in Part I, Section III, and it was noted that most of them would have to have large cross sections in order to significantly contribute to the poison or to alter the production rate of the nuclides they form by β^- decay. To the extent that these nuclides possess large cross sections they will alter the aggregate poison reported here, especially the calculated transients. Their medium half-life is about 70 hours. Therefore, even if each had a 6-percent yield, half would require a minimum cross section of 2000b in order to add ~1 percent to the late-in-life poison. Furthermore, the poison each would add would frequently be compensated by the reduction in poison of its radioactively formed daughter. The added progeny poison of the (n, γ) branch and the reduction in progeny poison of the radioactively formed daughter may, or may not, compensate. Thus the addition of cross sections for the 31 nuclides above may increase or decrease the aggregate poison. There are some cases like that of Pm-149 where the aggregate poison would not change at all since its progeny, except for short-lived (2.7h) Pm-150, are identical to the Sm-149 progeny. But however reassuring these comments may appear, about half of the above nuclides will ultimately require cross sectional measurements for there to be great confidence in calculations.

In summary, the unknown fission product poisons clearly constitute the main uncertainty in the calculations. This analysis has removed the uncertainty associated with coupling systematics, that is, with the analytical treatment. Uncertainty resulting from the measured yields and cross sections, and errors due to neglect of self-shielding and non- $1/v$ cross sections appear to be less than ± 10 percent. Therefore, it is believed that the present calculations are sufficiently accurate that a comparison with experimental measurements of the gross fission product poison in several flux environments and histories will determine whether there are significant unidentified poisons. One such comparison was made in the last section, and it indicates that no large contributor to the steady-state poison has been missed.

APPENDIX A

LINEAR CHAIN RESOLUTION

Consider a rather arbitrary branch point:

(chain 1A)

Let

y = yield fraction per fission

s = fission rate

λ = radioactive decay rate per nuclide

A = absorption rate per nuclide

$\alpha = \lambda + A$

Then, the coupled differential equations describing the concentration, N , of the nuclides are:

$$\dot{N}_1 = y_1 s - \alpha_1 N_1 \quad (1A)$$

$$\dot{N}_2 = y_2 s - \alpha_2 N_2 \quad (2A)$$

$$\dot{N}_3 = y_3 s + \lambda_1 N_1 + A_2 N_2 - \alpha_3 N_3 \quad (3A)$$

$$\dot{N}_4 = y_4 s + \lambda_3 N_3 - \alpha_4 N_4 \quad (4A)$$

$$\dot{N}_5 = y_5 s + A_3 N_3 - \alpha_5 N_5 \quad (5A)$$

where

$$\dot{N} = \frac{dN}{dt}$$

Consider N_3 ; since these are linear equations, N_3 can be divided into components: $N_3 = N_{3A} + N_{3B}$. Let N_{3A} be that part of N_3 due to the source term $\lambda_1 N_1$ and N_{3B} that due to the source $A_2 N_2$. The yield source, $y_3 s$, can be associated with either component or arbitrarily divided between N_{3A} and N_{3B} ; let it be a source to N_{3A} only. Similarly, N_4 can be divided into components; one, N_{4A} , due to the source $\lambda_3 N_{3A}$ and one, N_{4B} , due to the source $\lambda_3 N_{3B}$, and likewise for N_5 . Now there are four sets of coupled equations containing three "nuclides" each. Their equations and the associated linear chains that each represents are:

$$\dot{N}_1 = y_1 s - \alpha_1 N_1$$

$$\dot{N}_{3A} = y_3 s + \lambda_1 N_1 - \alpha_3 N_{3A}$$

$$\dot{N}_{4A} = y_4 s + \lambda_3 N_{3A} - \alpha_4 N_{4A}$$

$$\dot{N}_2 = y_2 s - \alpha_2 N_2$$

$$\dot{N}_{3B} = A_2 N_2 - \alpha_3 N_{3B}$$

$$\dot{N}_{4B} = \lambda_3 N_{3B} - \alpha_4 N_{4B}$$

$$\dot{N}_1 = y_1 s - \alpha_1 N_1$$

$$\dot{N}_{3A} = y_3 s + \lambda_1 N_1 - \alpha_3 N_{3A}$$

$$\dot{N}_{5A} = y_5 s + A_3 N_{3A} - \alpha_5 N_{5A}$$

$$\dot{N}_2 = y_2 s - \alpha_2 N_2$$

$$\dot{N}_{3B} = A_2 N_2 - \alpha_3 N_{3B}$$

$$\dot{N}_{5B} = A_3 N_{3B} - \alpha_5 N_{5B}$$

Each set of equations can now be solved independently of the other sets. All nuclides labeled with the same number but with a different letter are then added (once) to get the total concentration of the nuclide, e.g., $N_3 = N_{3A} + N_{3B}$. N_{3A} can be obtained from either chain 2A or 4A, whereas N_{3B} can be obtained from either chain 3A or 5A, etc. There is, of course, considerable redundancy. Some nuclide and partial nuclide concentrations have been calculated twice; however, the calculational effort per set is reduced. Also, there is a considerable conceptual gain since all chains now involve the simplest possible coupling sequence, and their formation is rather intuitive. Nuclide concentrations derived from various sources are quantitatively given. Of more importance, each set of equations has the same general form, and a general solution applies to all sets.

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APPENDIX B

GENERAL SOLUTION FOR THE N-NUCLIDE CHAIN

The differential equations for n coupled nuclides formed into a linear chain can be written in terms of previously defined quantities:

$$\begin{aligned}
 \frac{dN_1}{dt} &= \overline{Y_1 S} - (\lambda_1 + A_1) N_1 \\
 \frac{dN_2}{dt} &= \overline{Y_2 S} + \gamma_1 N_1 - (\lambda_2 + A_2) N_2 \\
 \frac{dN_n}{dt} &= \overline{Y_n S} + \gamma_{n-1} N_{n-1} - (\lambda_n + A_n) N_n
 \end{aligned}
 \tag{1-B}$$

These equations are readily converted into a set of linear algebraic equation by the Laplace transform, assuming constant coefficients. For simplicity the following definitions are used.

$$\beta_i = s + \lambda_i + A_i \tag{2-B}$$

$$C_i = \frac{\overline{Y_i S}}{s} + N_i(0)$$

where s = transform variable.

After conversion the equations can be written in matrix form:

$$\begin{pmatrix}
 \beta_1 & & & & \\
 -\gamma_1 & \beta_2 & & & \\
 & -\gamma_2 & \beta_3 & & \\
 & & & \ddots & \\
 & & & & \beta_{n-1} \\
 & & 0 & & -\gamma_{n-1} \beta_n
 \end{pmatrix}
 \begin{pmatrix}
 N_1 \\
 N_2 \\
 \vdots \\
 \vdots \\
 N_n
 \end{pmatrix}
 =
 \begin{pmatrix}
 C_1 \\
 C_2 \\
 \vdots \\
 \vdots \\
 C_n
 \end{pmatrix}
 \tag{3-B}$$

This is solved for $N_n(s)$ by Cramer's rule. The determinant of the matrix is clearly $\beta_1 \beta_2 \dots \beta_n$. The determinant obtained on replacing the n^{th} column by (C_i) is

$$\begin{aligned}
 &\gamma_1 \gamma_2 \dots \gamma_{n-1} C_1 + \beta_1 \gamma_2 \gamma_3 \dots \gamma_{n-1} C_2 + \beta_1 \beta_2 \gamma_3 \gamma_4 \dots \gamma_{n-1} C_3 + \dots \\
 &+ \beta_1 \beta_2 \dots \beta_{n-2} \gamma_{n-1} C_{n-1} + \beta_1 \beta_2 \dots \beta_{n-1} C_n
 \end{aligned}$$

Hence the solution in terms of the transform variable is

$$N_n(s) = \frac{C_n}{\beta_n} + \frac{\gamma_{n-1} C_{n-1}}{\beta_{n-1} \beta_n} + \frac{\gamma_{n-2} \gamma_{n-1} C_{n-2}}{\beta_{n-2} \beta_{n-1} \beta_n} + \dots + \frac{\gamma_1 \gamma_2 \dots \gamma_{n-1} C_1}{\beta_1 \beta_2 \dots \beta_n} \quad 4-B$$

Or, in terms of the general term of Equation (4-B),

$$\frac{\gamma_m \gamma_{m+1} \dots \gamma_{n-1} C_m}{\beta_m \beta_{m+1} \dots \beta_n},$$

Equation (4-A) becomes

$$N_n(s) = \sum_{m=1}^n \frac{\gamma_m \gamma_{m+1} \dots \gamma_{n-1} C_m}{\beta_m \beta_{m+1} \dots \beta_n} \quad 5-B$$

Converting to the previously defined quantities, Equations (2-B),

$$N_n(s) = \sum_{m=1}^n \frac{\left(\frac{\overline{Y_m S}}{s} + N_m(o) \right) \frac{1}{\gamma_n} \prod_{k=m}^n \gamma_k}{\prod_{i=m}^n (s + \lambda_i + A_i)} \quad 6-B$$

Using partial fractions this may be expanded and inverted to yield the final solution:

$$N_n(t) = \sum_{m=1}^n \frac{1}{\gamma_n} \prod_{k=m}^n \gamma_k \left\{ \frac{\overline{Y_m S}}{s} \left[\frac{1}{\prod_{l=m}^n (\lambda_l + A_l)} \right] - \sum_{j=m}^n \frac{\exp [-(\lambda_j + A_j)t]}{(\lambda_j + A_j) \prod_{\substack{i=m \\ i \neq j}}^n (\lambda_i + A_i - \lambda_j - A_j)} \right\} + N_m(o) \left(\sum_{j=m}^n \frac{\exp [-(\lambda_j + A_j)t]}{\prod_{\substack{i=m \\ i \neq j}}^n (\lambda_i + A_i - \lambda_j - A_j)} \right) \quad 7-B$$

This solution assumes that $(\lambda_i + A_i) \neq (\lambda_j + A_j)$ for $i \neq j$ and that $(\lambda + A) \neq 0$. It is readily converted to the form of Equation (2).

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